

Computational Nanoscience at NERSC

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Lawrence Berkeley National Lab**

**US Department of Energy
Office of Science**

- What can we do ?
- How do we do it ?
- Examples

Material as a mark of civilization

Stone age



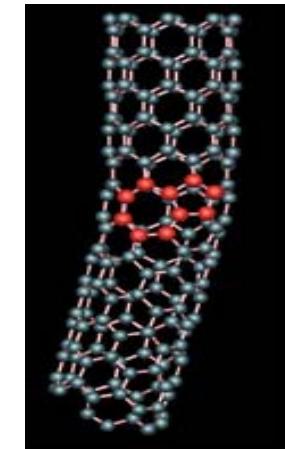
Bronze age



Semiconductor information age



Nanostructure age

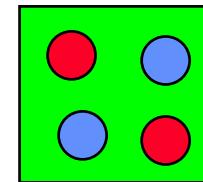


Nanoscience is a material science:

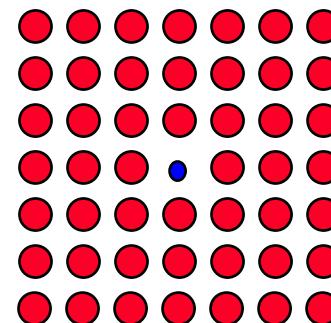
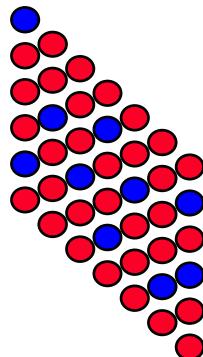
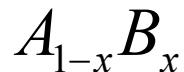
Nano size building block
Assemble them into device

Making new solid state materials

- New crystal compounds

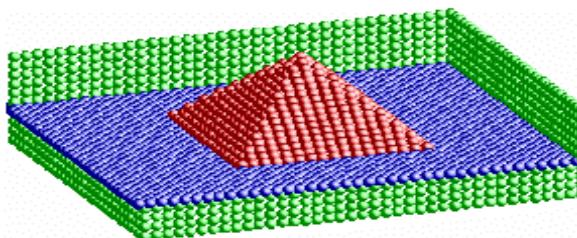


- Alloys



- Impurity and doping

- Modifying the size and shape of the material

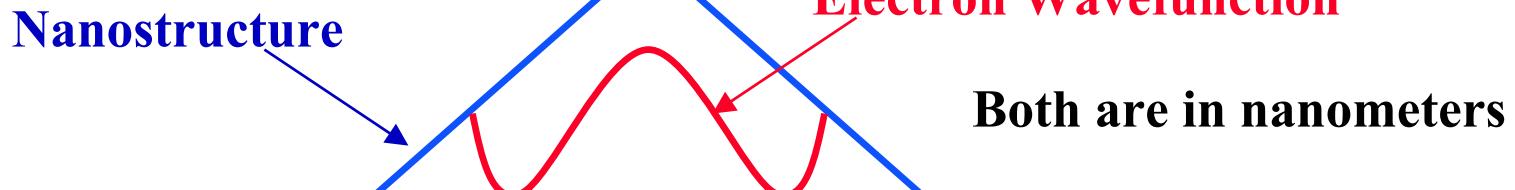
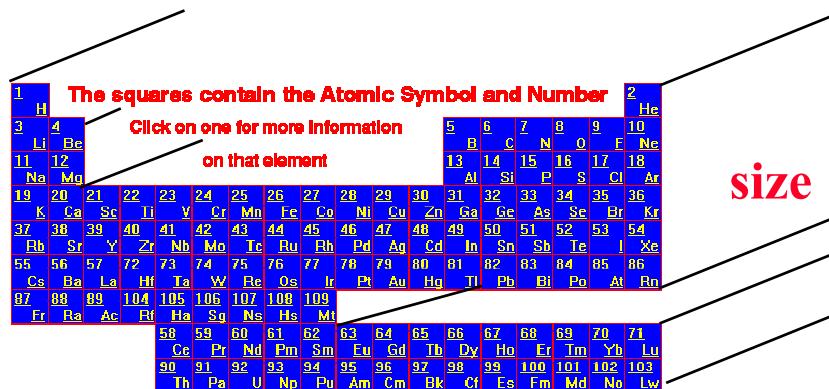


Nanostructure as a new material

Definition: Nanostructure is an assembly of nanometer scale “building blocks”.



Why nanometer scale: This is the scale when the properties of these “building blocks” become different from bulk.



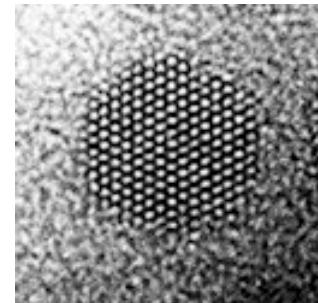
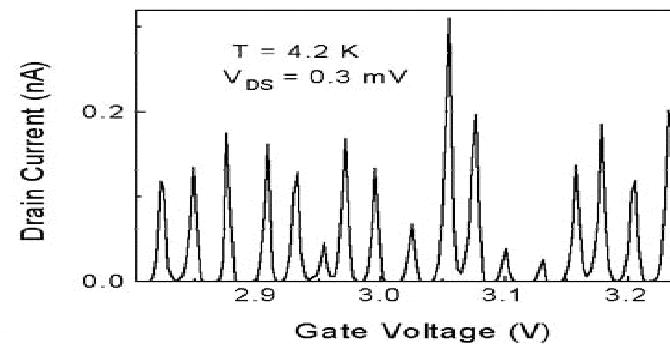
Examples of new properties

- Band gap increase



CdSe quantum dot

- Single electron effects on transport (Coulomb blockade).



- Mechanical properties, surface effects and no dislocations

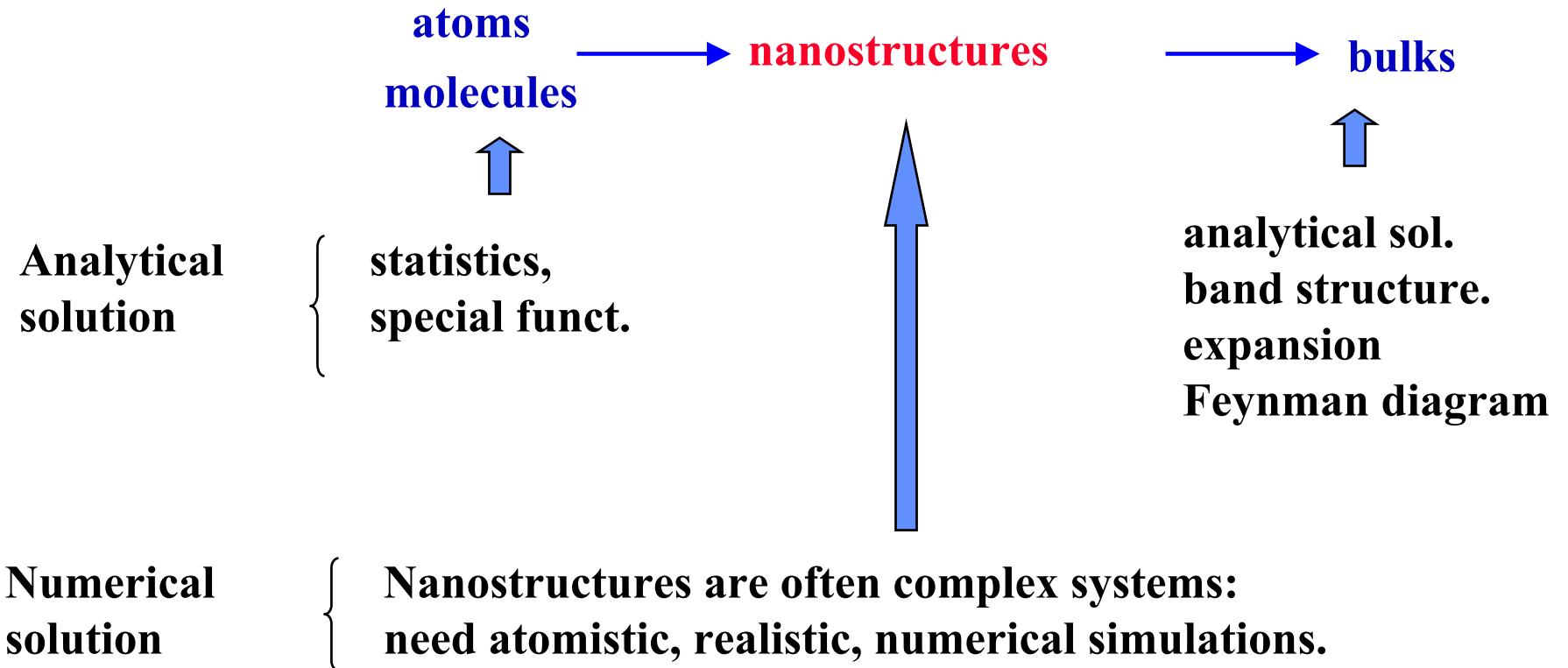
Theoretical Challenge

Three corner stones of modern science:

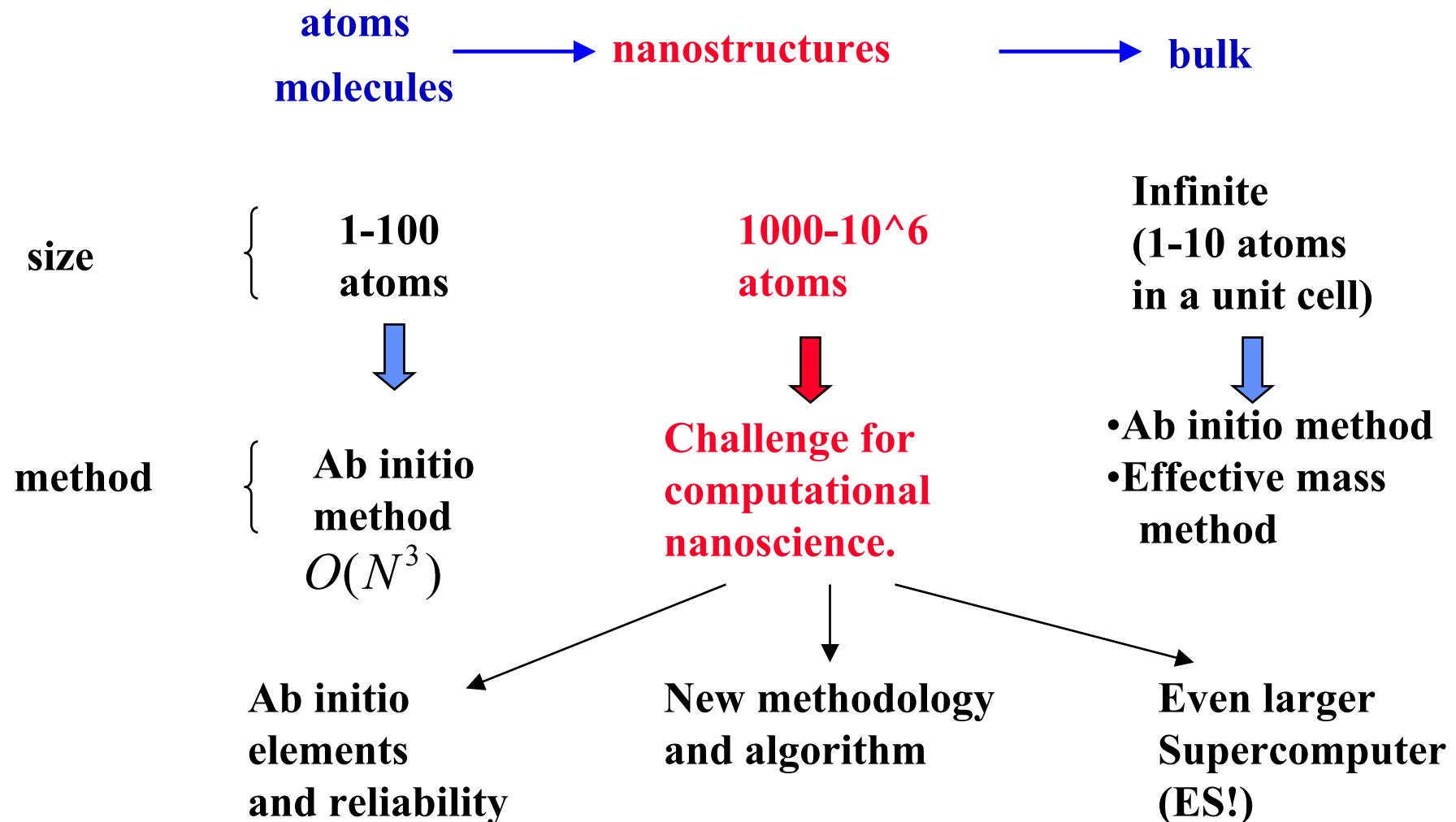
Experiment

Theoretical analysis

Computational simulation



Computational challenge



Ab initio electronic structure calculations

All the material science problems are solved !

---- Schroedinger, 1930's

$$\left\{ -\sum_i \frac{1}{2} \nabla_i^2 + \sum_{i,j} \frac{1}{|r_i - r_j|} + \sum_{i,R} \frac{Z}{|r_i - R|} \right\} \Psi(r_1,..r_N) = E \Psi(r_1,..r_N)$$

Linear equation, but extremely large dimension: $\Psi(r_1,..r_2) N^N$

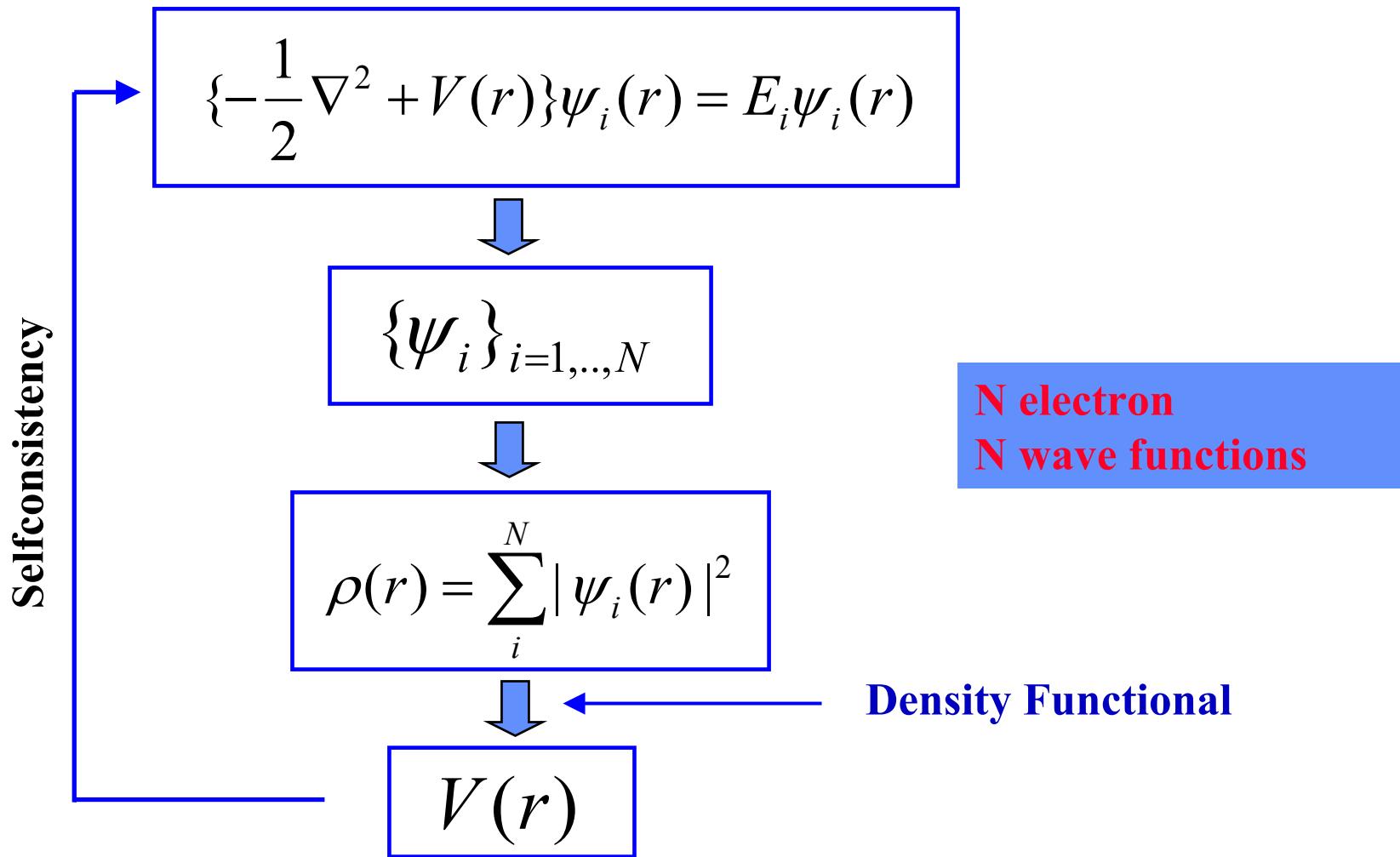
Density functional theory and local density approximation

---- W. Kohn's 1997 Nobel prize

$$\left\{ -\frac{1}{2} \nabla^2 + V(r, [\rho(r)]) + \sum_R \frac{1}{|r - R|} \right\} \psi_i(r) = E_i \psi_i(r) \quad N^2$$

$$\rho(r) = \sum_i |\psi_i(r)|^2 \qquad \qquad \psi_i(r) : \text{single electron wave function}$$

Ab initio density functional calculations



Two tasks for a hybrid nano computation method

(1) To get the potential $V(r)$ [or the charge density $\rho(r)$]

so we will have the Hamiltonian.

(We want ab initio reliability, but not a full ab initio calculation)

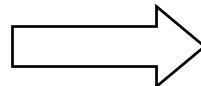
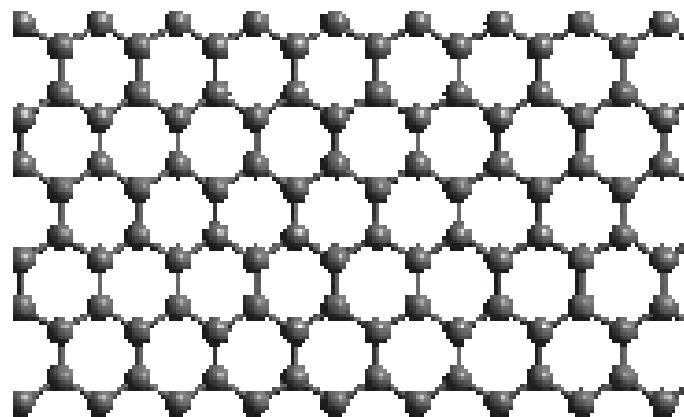
(2) To solve the single particle Hamiltonian
(Schroedinger's equation), to get the physical properties.

$$\left\{-\frac{1}{2}\nabla^2 + V(r)\right\}\psi_i(r) = E_i\psi_i(r)$$

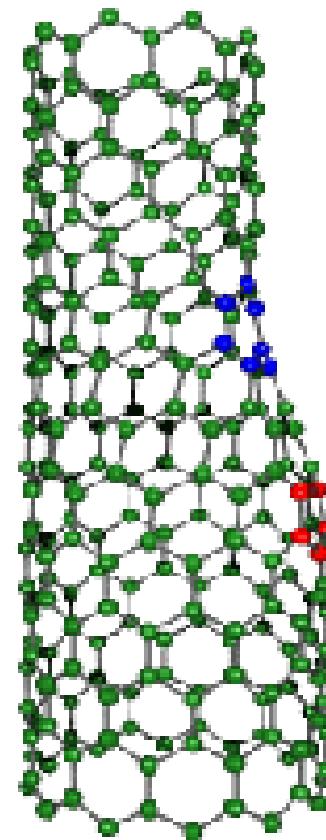
(Not the usual PDE, many eigen states, don't want and need to solve all of them)

Charge patching method

**Selfconsistent LDA
calculation of a single
graphite sheet**

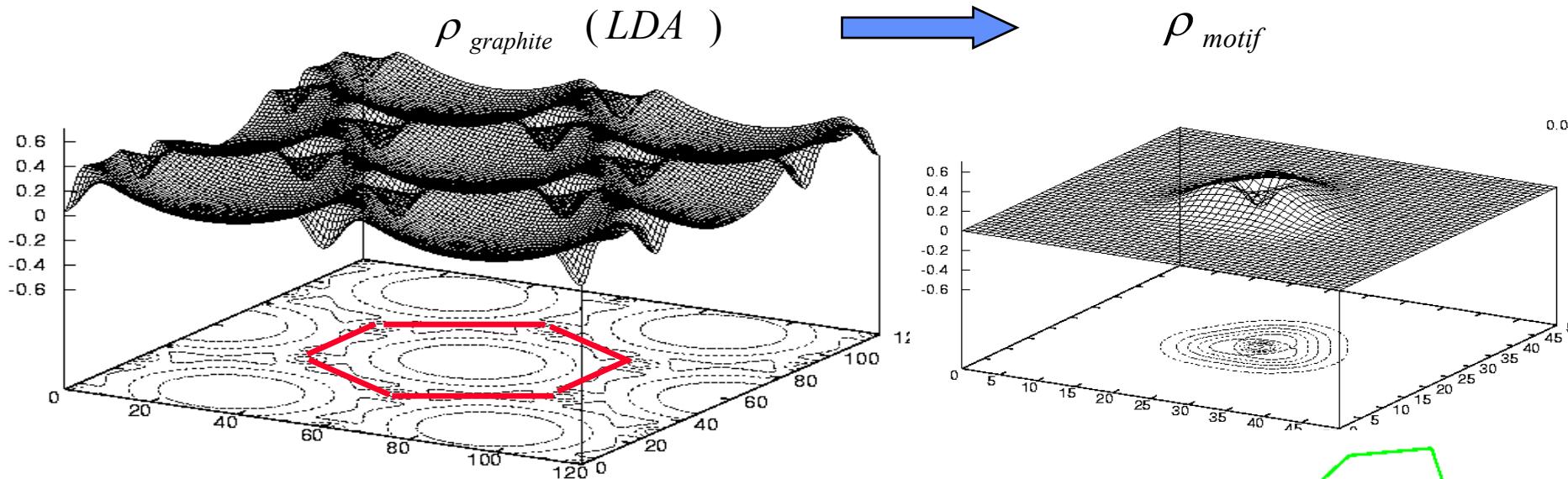


**Non-selfconsistent LDA
quality potential for
nanotube**



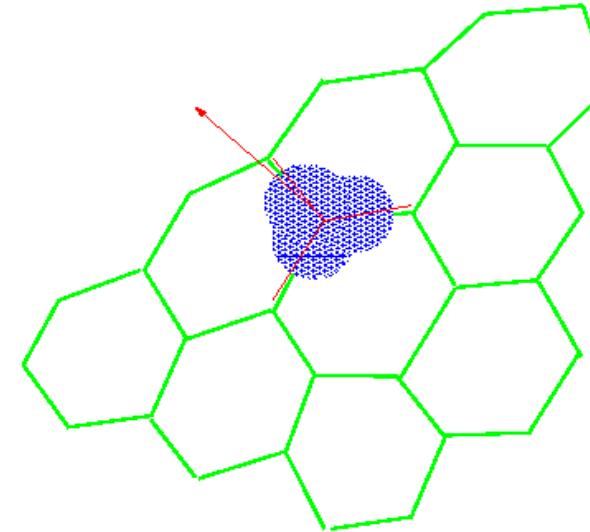
**Get information from small
system ab initio calc., then generate
the charge densities for large systems**

Motif based charge patching method



$$\rho_{nanotube}^{patch}(r) = \sum_R \rho_{motif}^{aligned}(r - R)$$

Error: 1%, ~20 meV eigen energy error.



Charge patching: free standing quantum dots

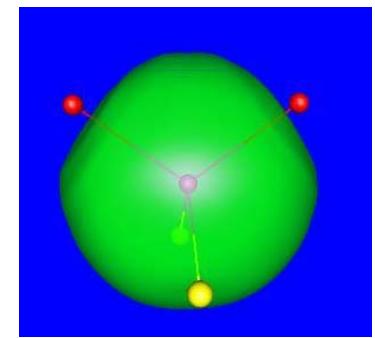
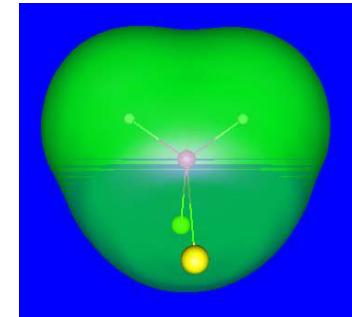
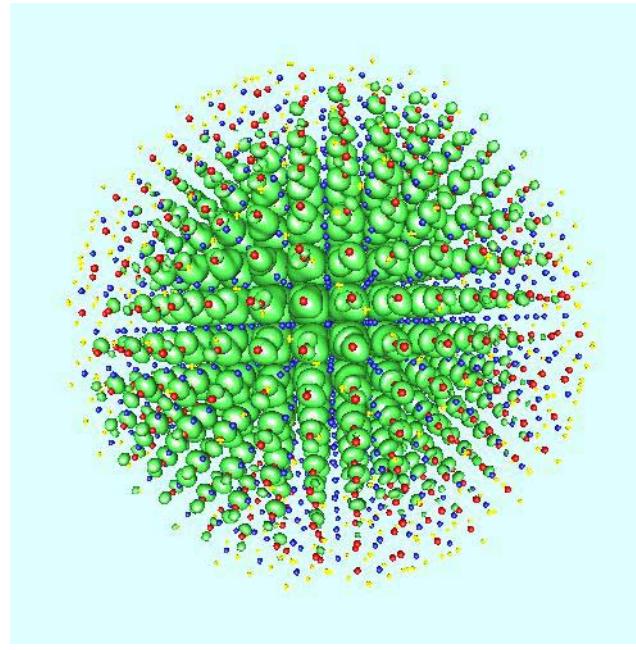
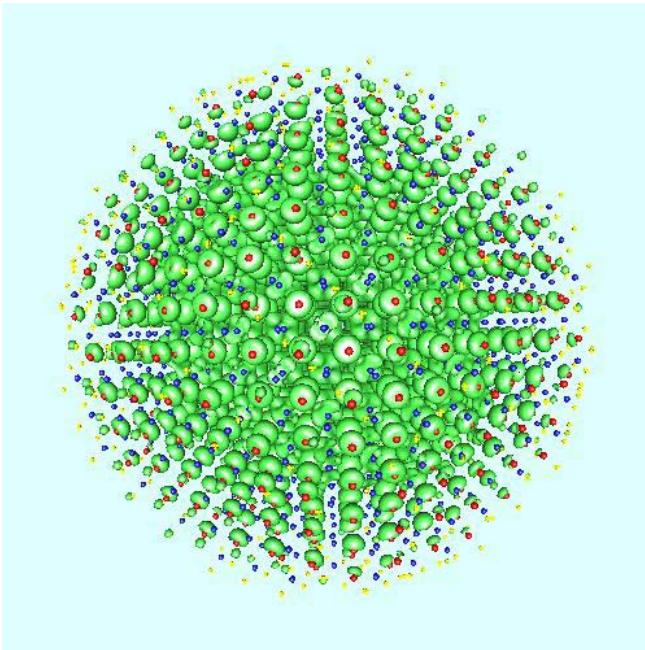
$\text{In}_{675}\text{P}_{652}$ LDA quality calculations (eigen energy error ~ 20 meV)

64 processors (IBM SP3) for ~ 1 hour

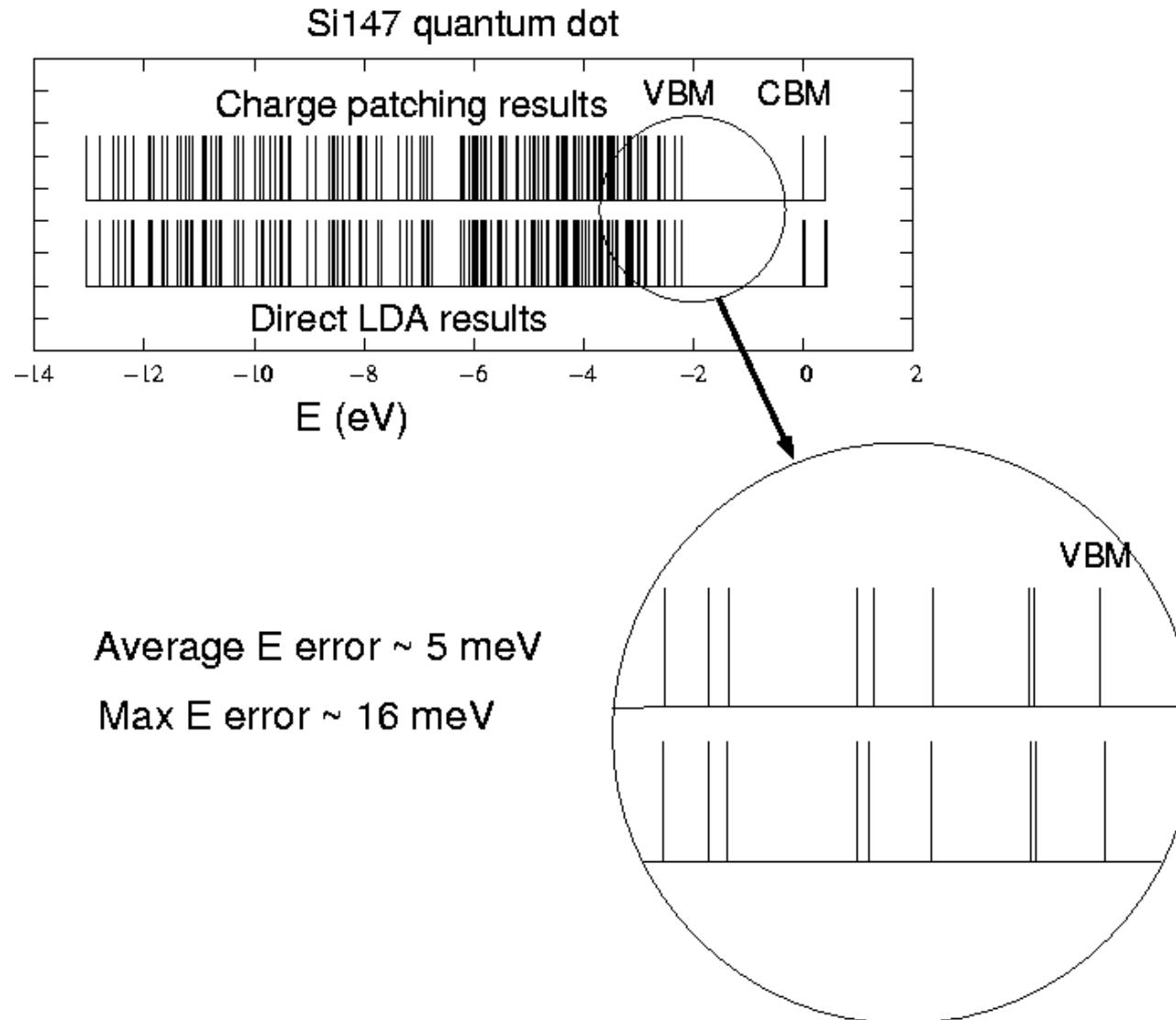
CBM

VBM

Total charge density motifs



The accuracy for the small Si quantum dot

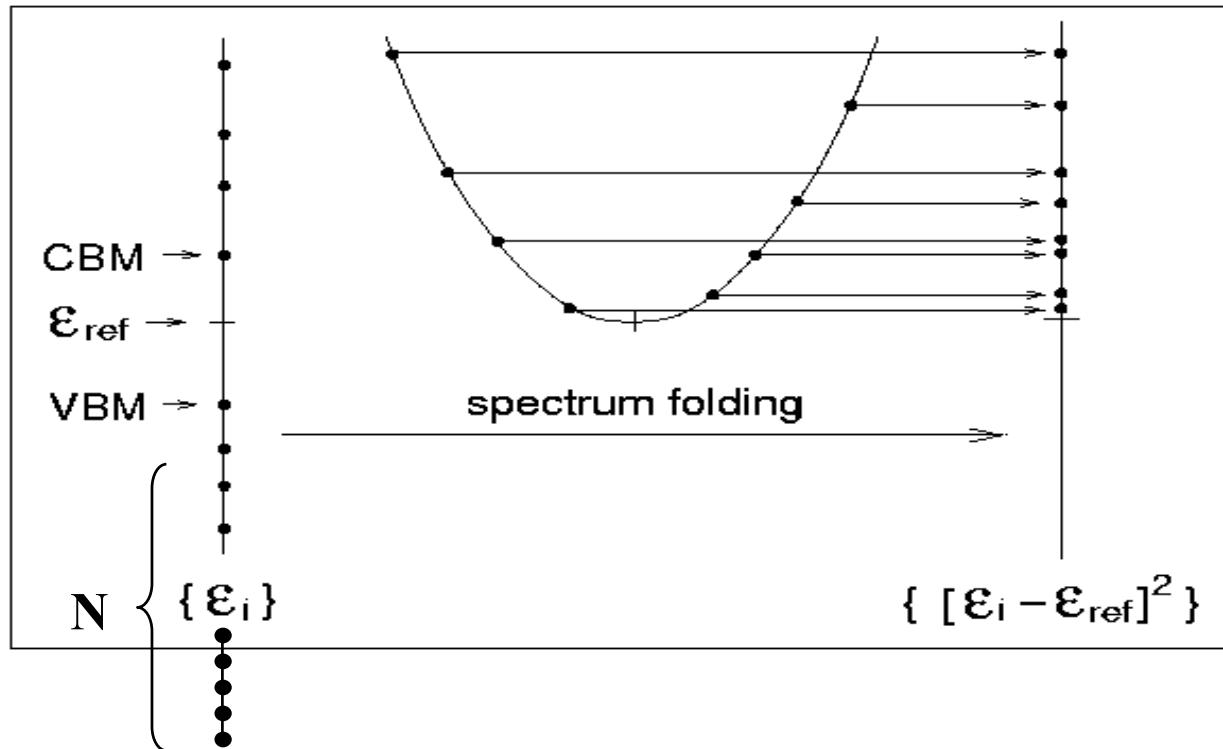


Folded Spectrum Method

$$\left\{-\frac{1}{2}\nabla^2 + V(r)\right\}\psi_i(r) = E_i\psi_i(r)$$

$$H\psi_i = \varepsilon_i\psi_i$$

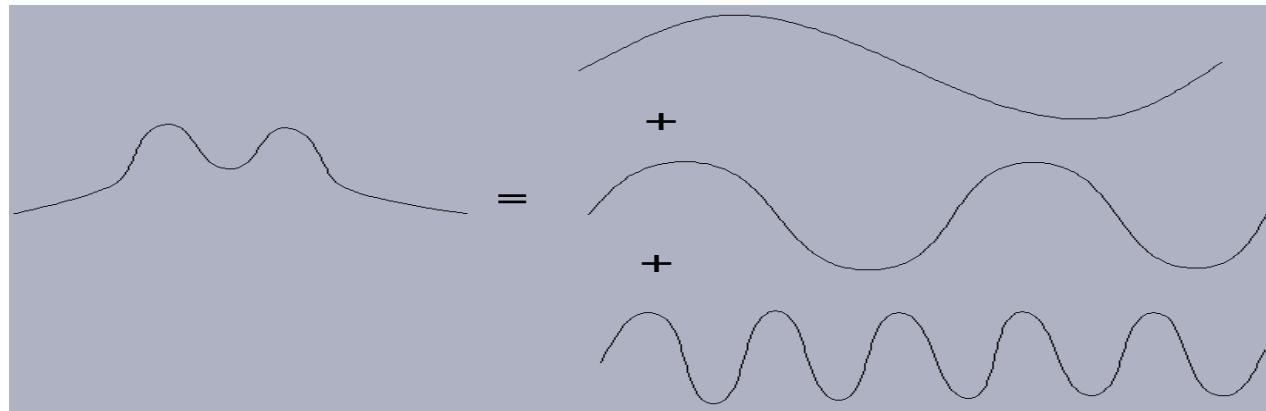
$$(H - \varepsilon_{ref})^2\psi_i = (\varepsilon_i - \varepsilon_{ref})^2\psi_i$$



Planewave expansion of the wavefunction

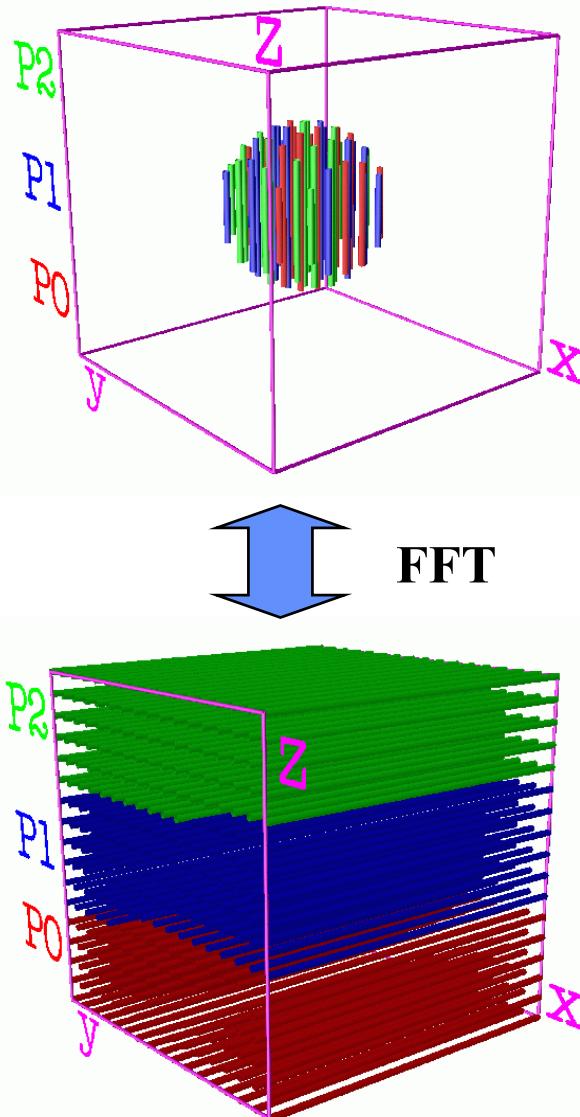
$$\left\{ -\frac{1}{2} \nabla^2 + V(r) \right\} \psi_i(r) = E_i \psi_i(r)$$

$$\psi(r) = \sum_q C(q) e^{iqr}$$

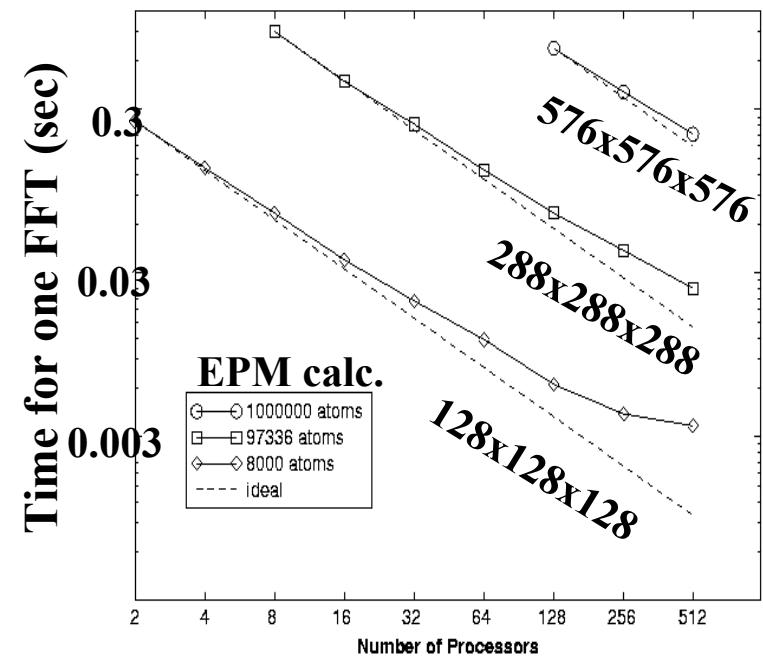


**Fast Fourier Transformation between
real space $\psi(r)$ and Fourier space $C(q)$.**

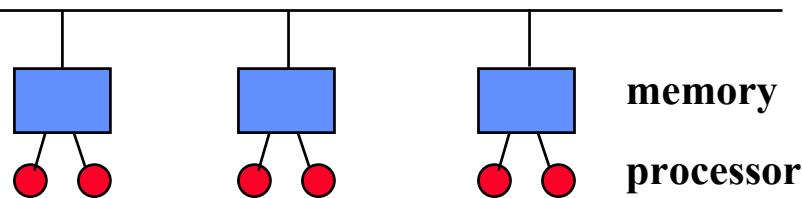
A parallel Fast Fourier Transformation code



- Specially designed for PW elec. structure calculation.
- Work load balance
- Memory balance
- Minimum communication

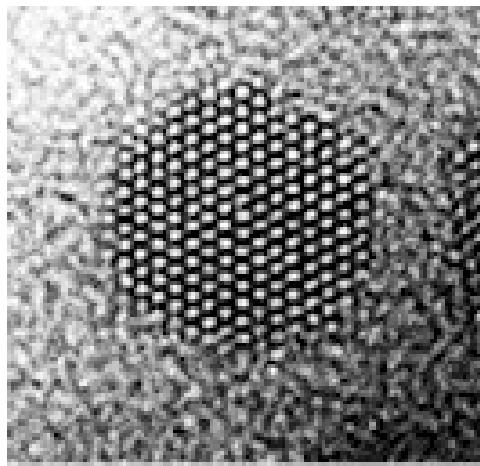


NERSC: National Energy Research Scientific Computing Center



6000 IBM SP3 processors, total peak speed: ~ 5 Tflop

Free standing quantum dots



CdSe
quantum dot
TEM image

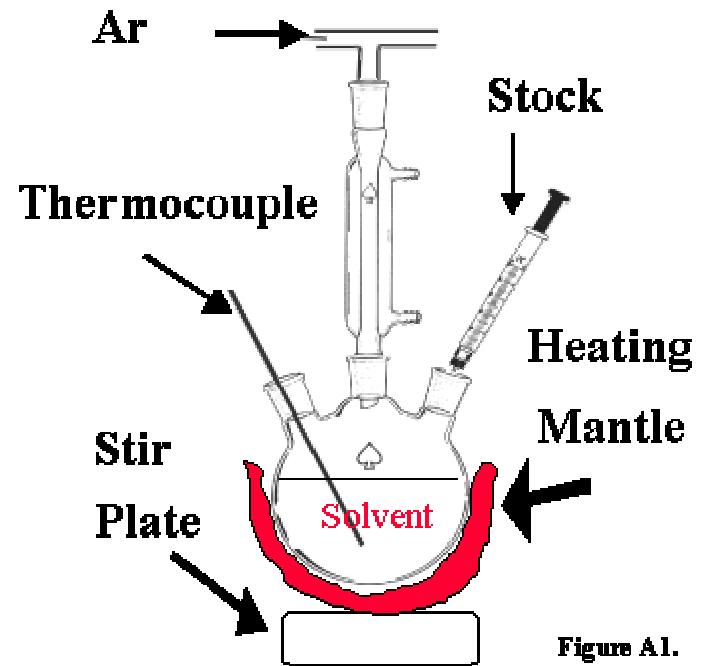
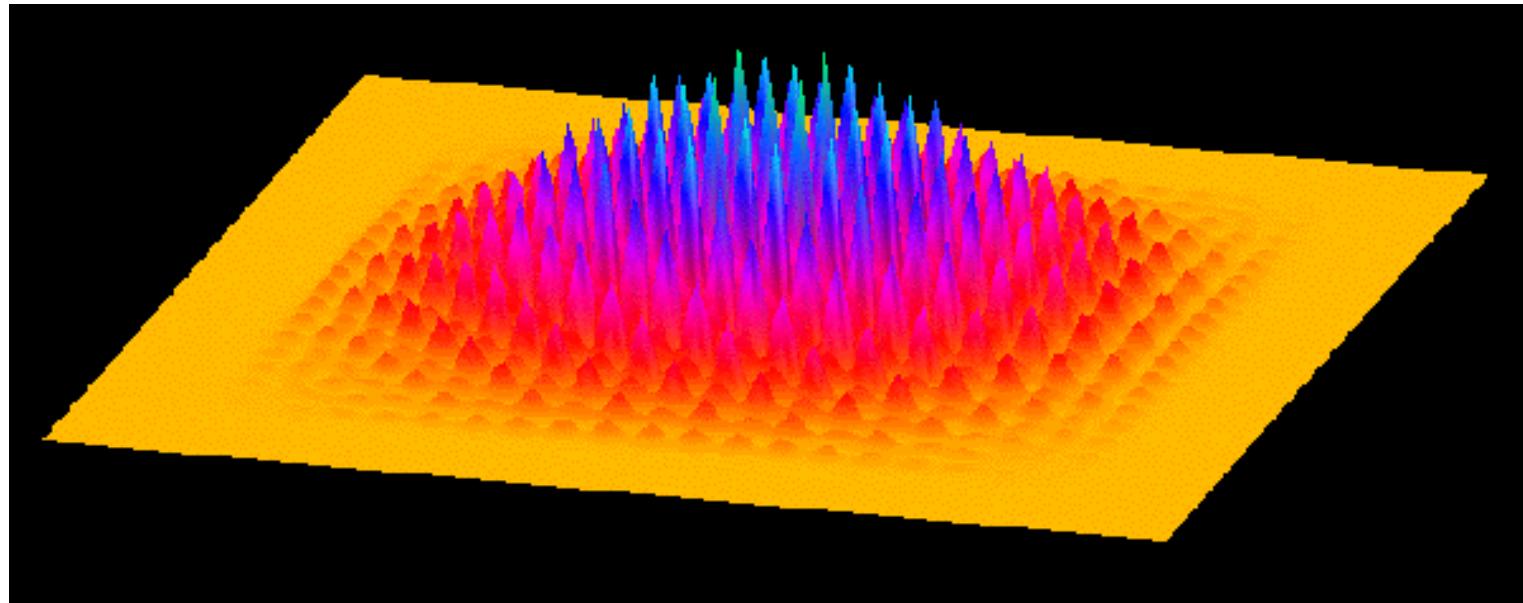


Figure A1.

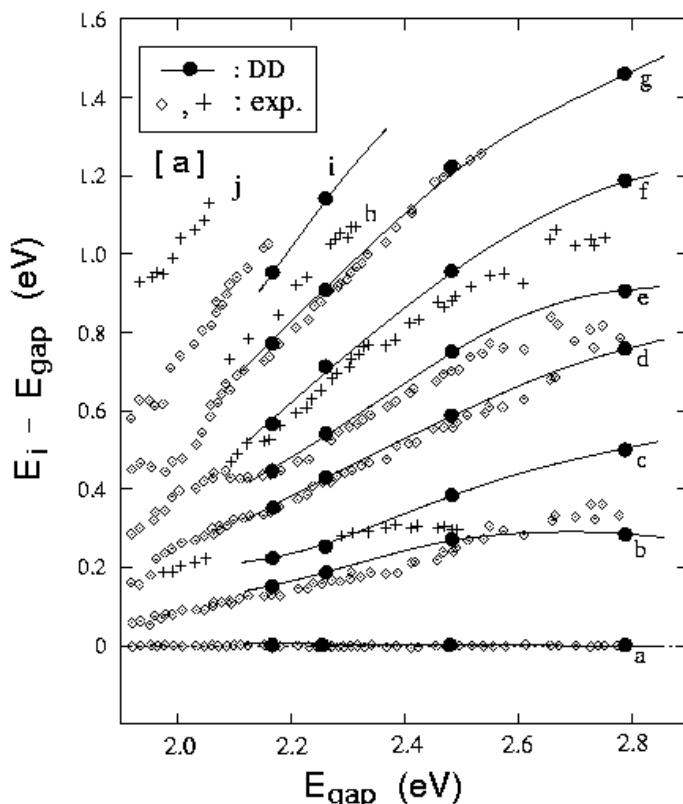
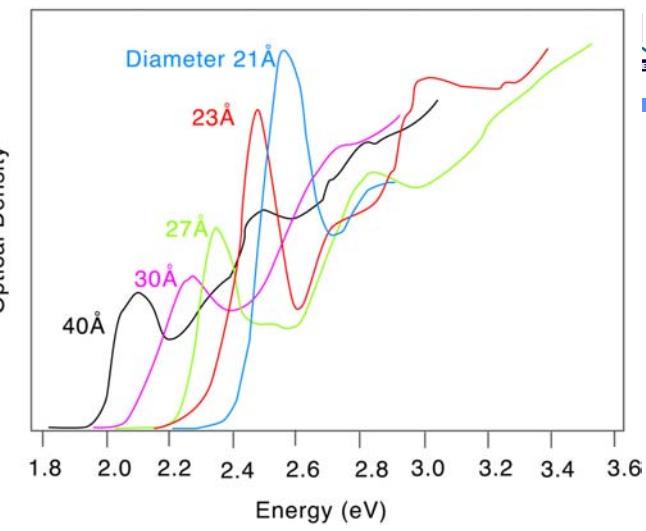
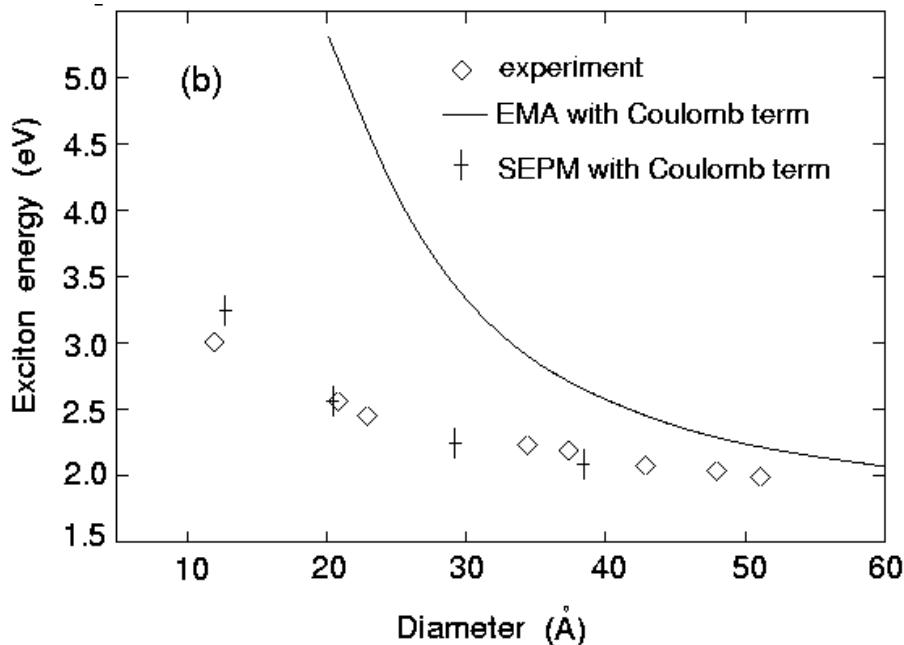
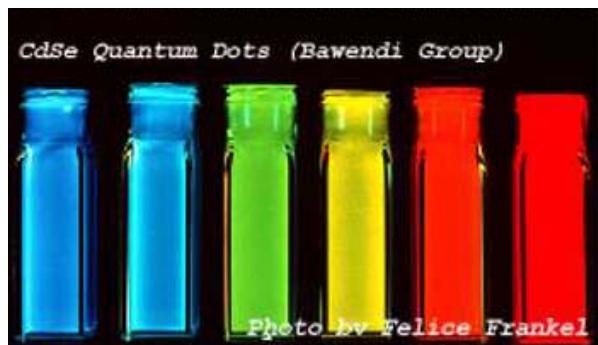
- Chemically synthesised
- Interior atoms are in bulk crystal structure
- Surface atoms are passivated
- Diameter ~ 20-100 Å
- A few thousand atoms, beyond ab initio method

Quantum dot wavefunctions

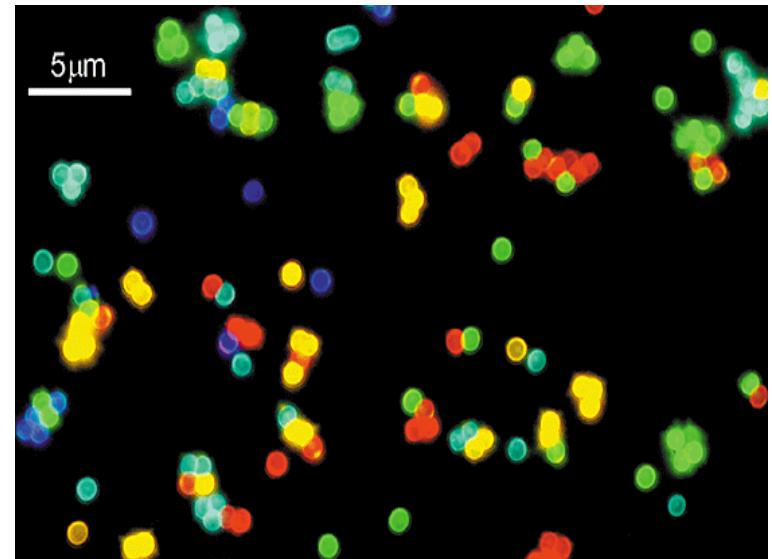
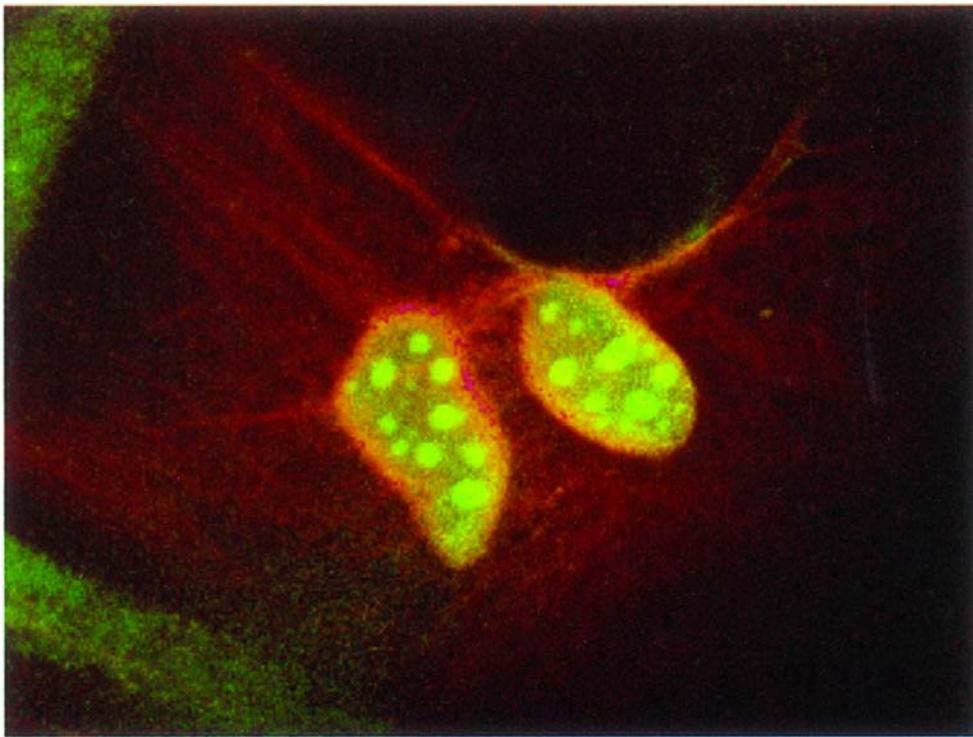


Cross section electron wavefunctions

CdSe quantum dot results

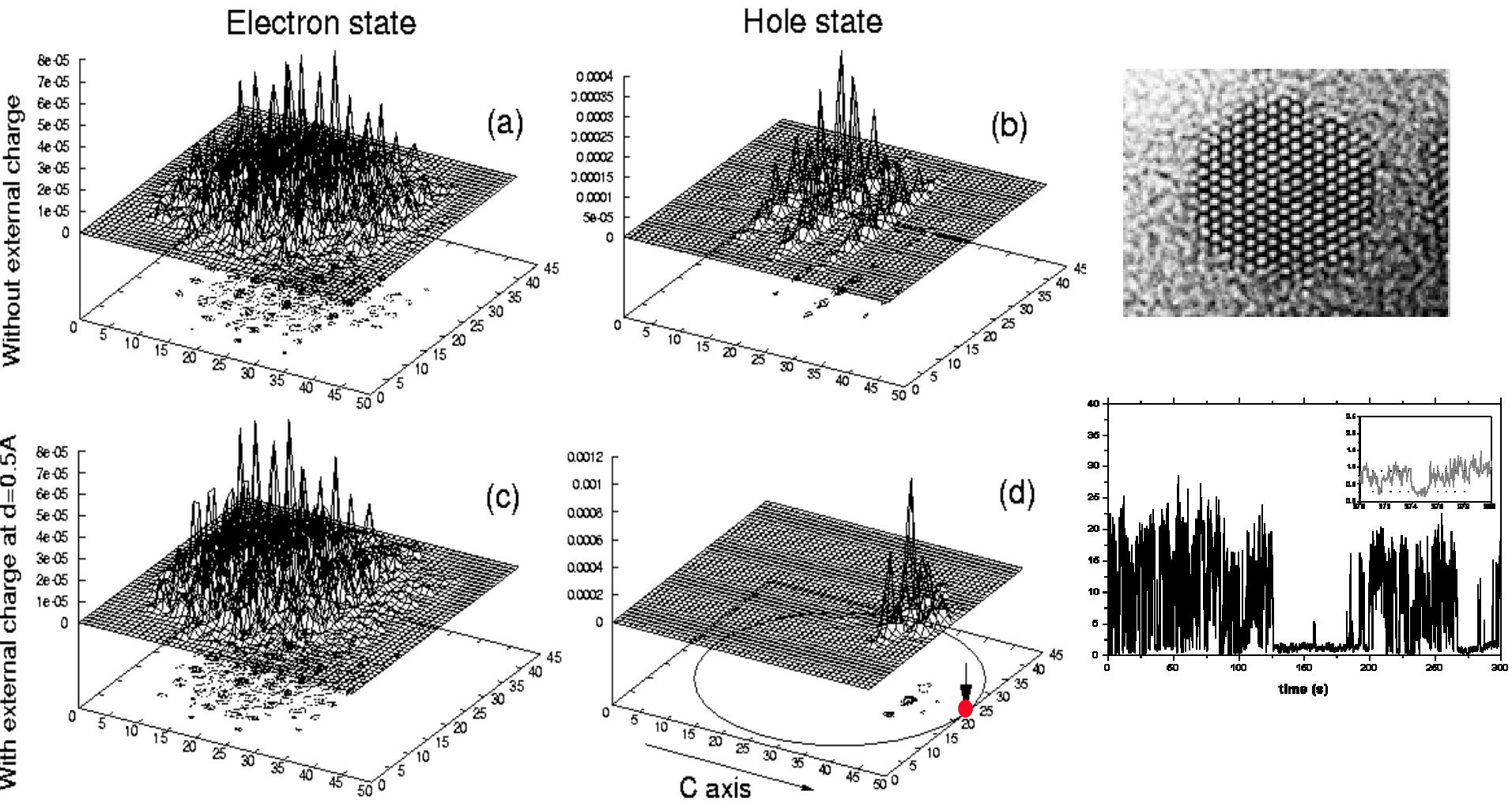


CdSe quantum dots as biological tags

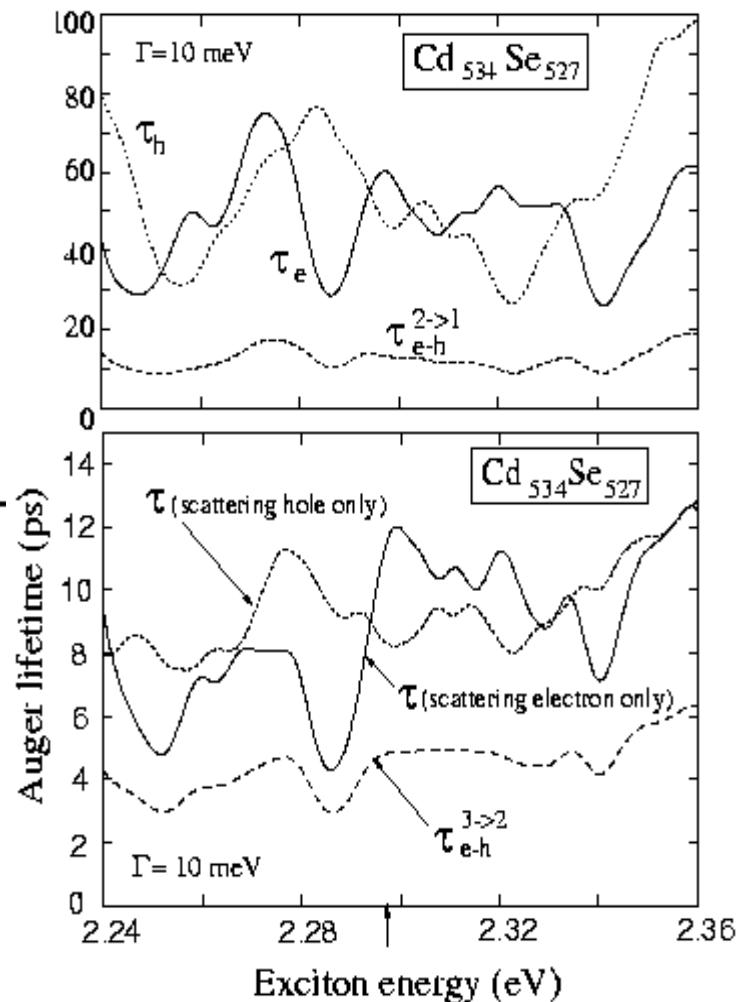
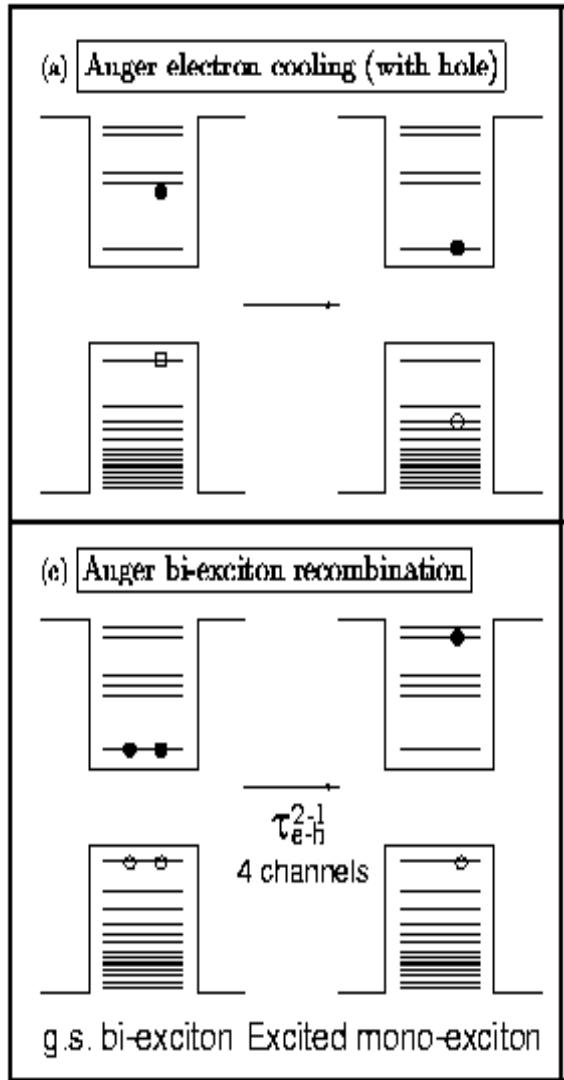


- Optically more stable than dye molecules
- Can have multiple colors

Photoluminescence intermittency of CdSe QD



Auger effect in CdSe quantum dot



Auger life times

Exp.

Calc.

Cooling

>0.5ps ~0.2-0.5ps

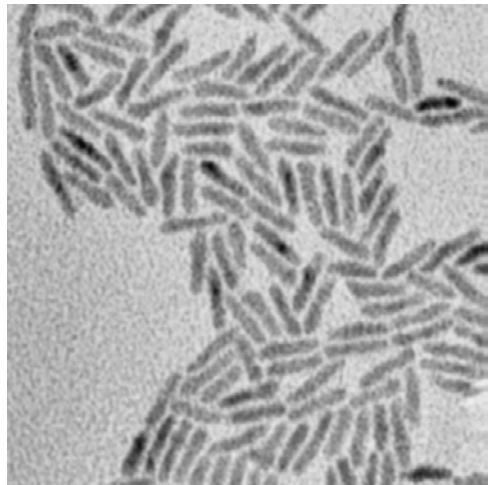
2 exciton->1 exc.

~2.7 ps ~2. ps

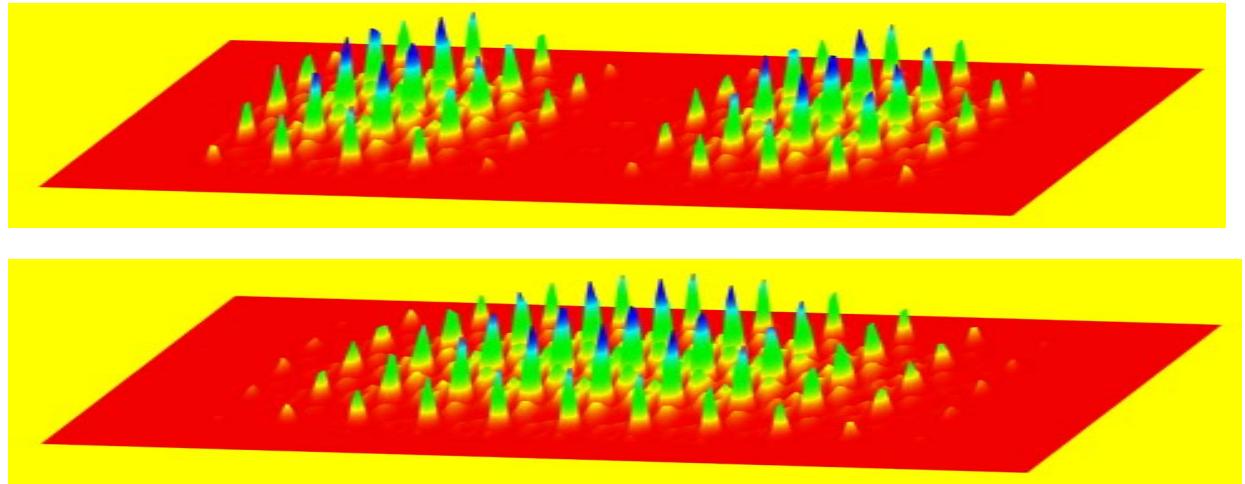
$\tau_{2eh \rightarrow 1eh} / \tau_{3eh \rightarrow 1eh}$

2.7 2.4

Polarization of CdSe quantum rods

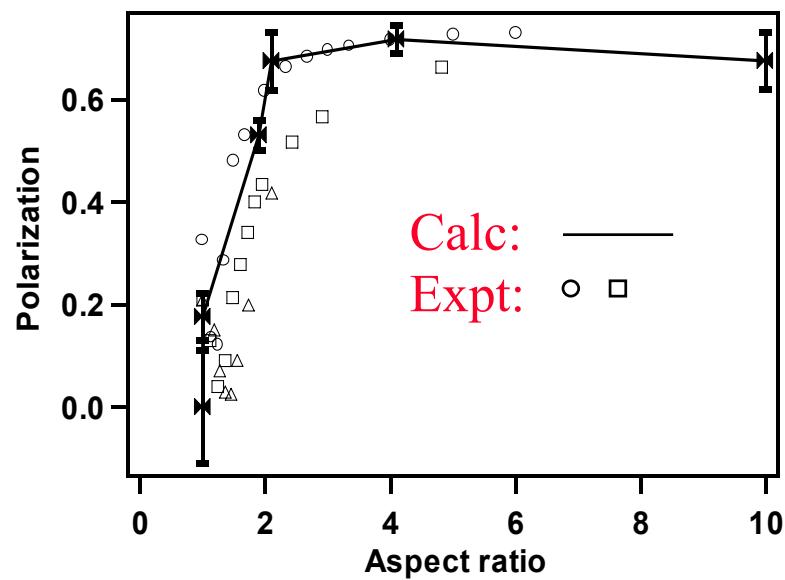
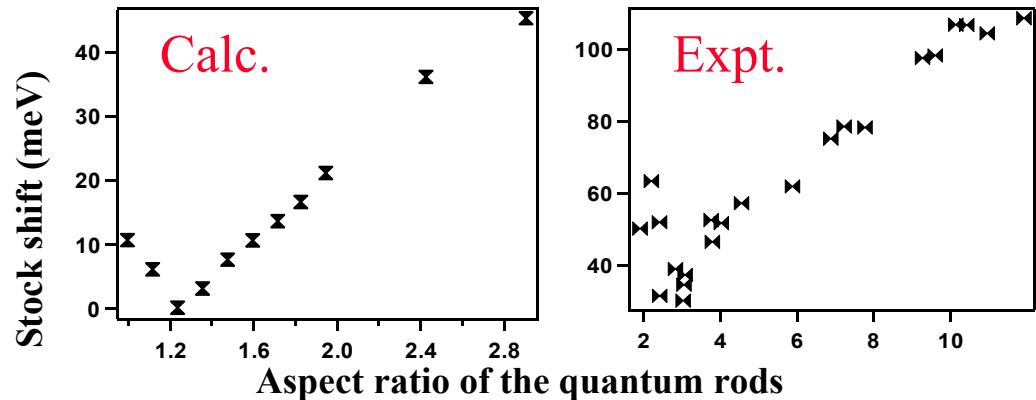
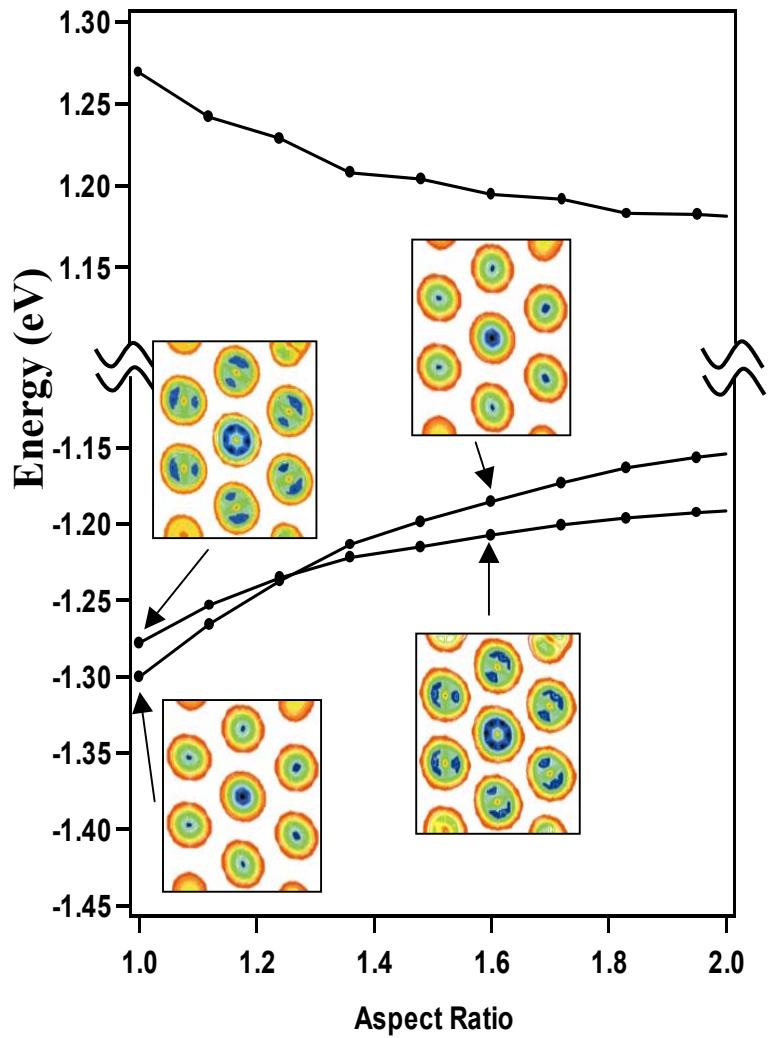


CdSe quantum rods

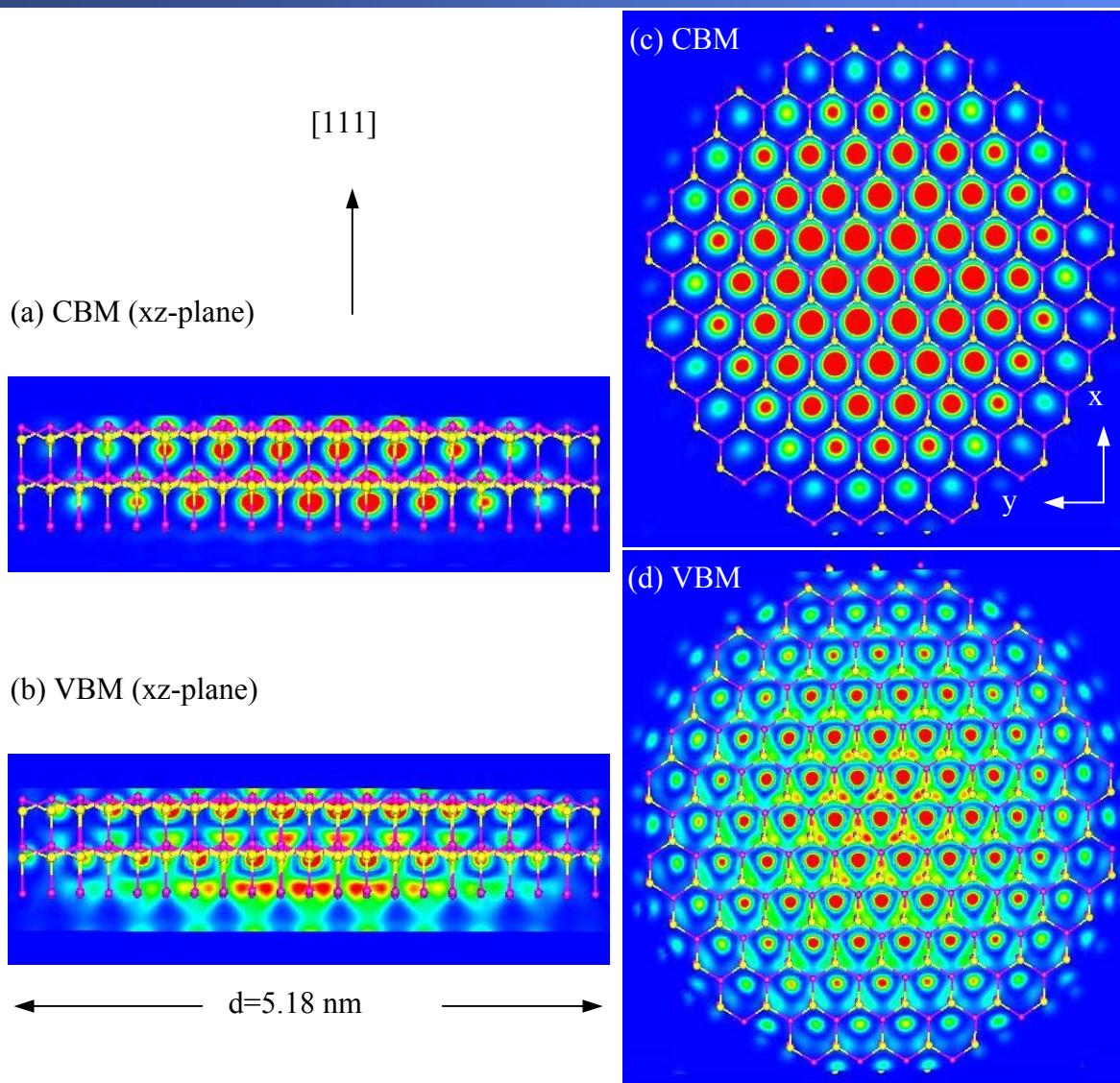


The electron wavefunctions of a quantum rods

Polarization of quantum rods (continued)

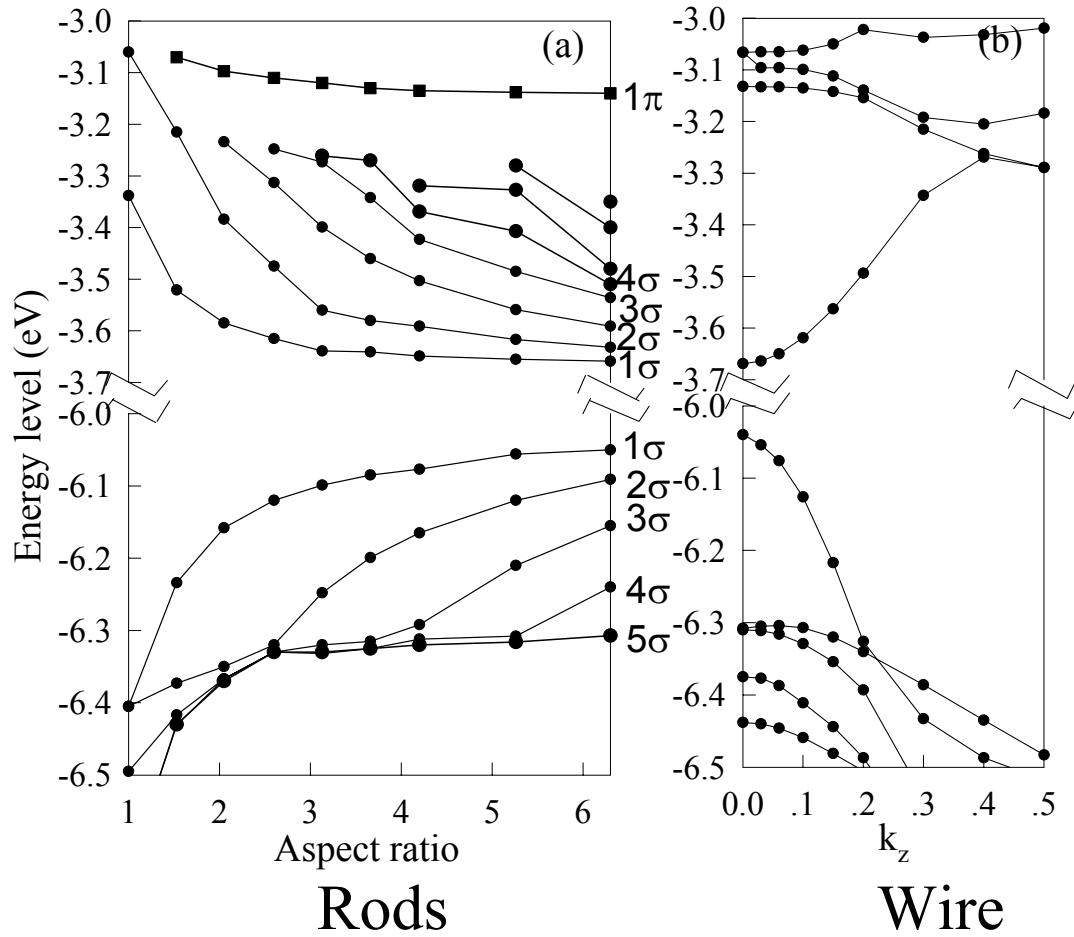


Quantum wire electronic states

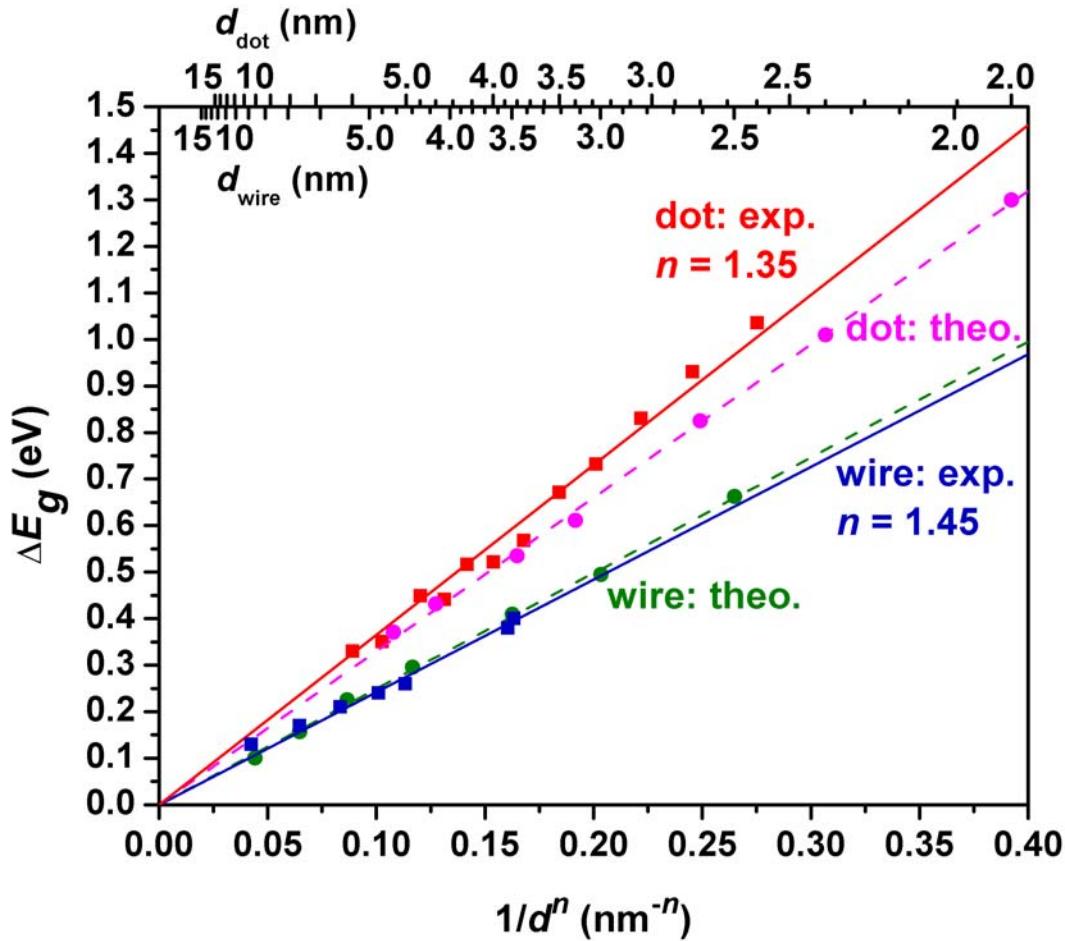


InP quantum rods and wires

(111) direction rods and wires



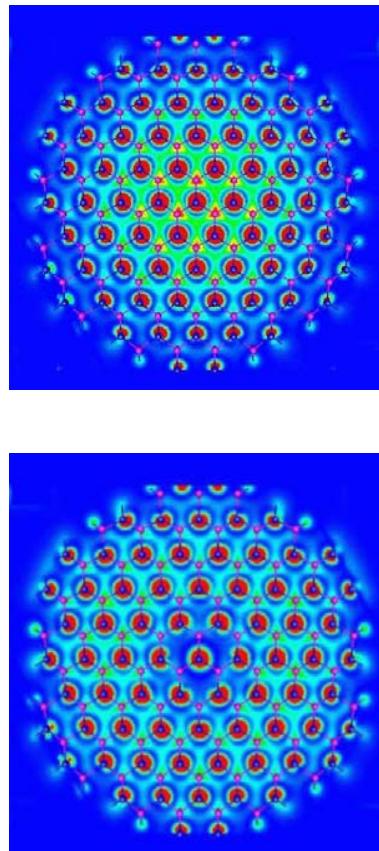
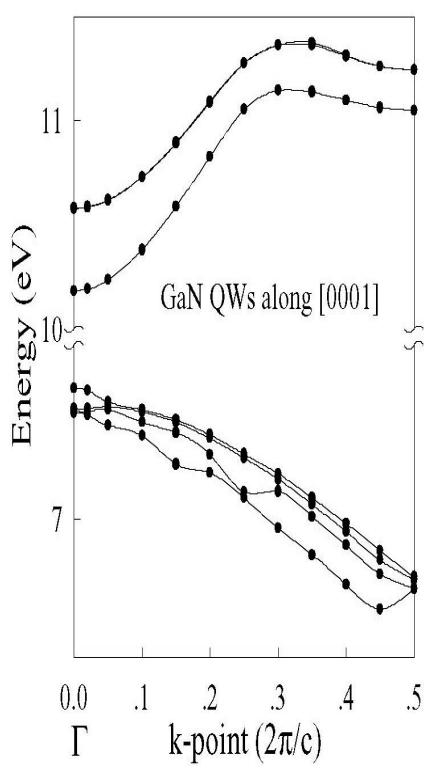
InP wires / InP dots



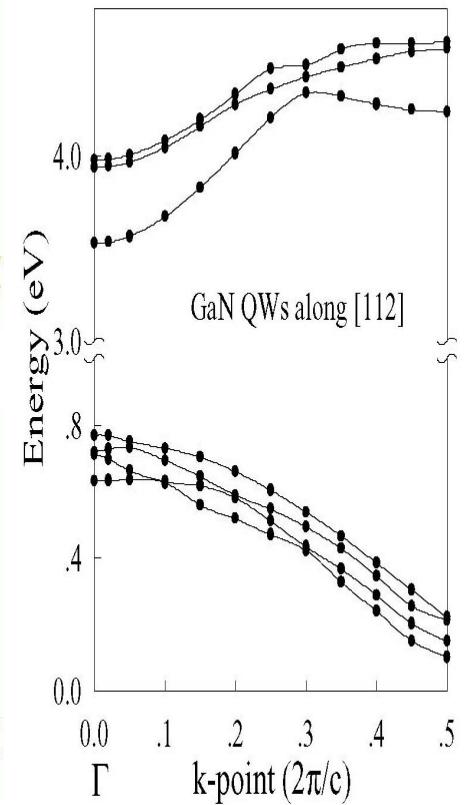
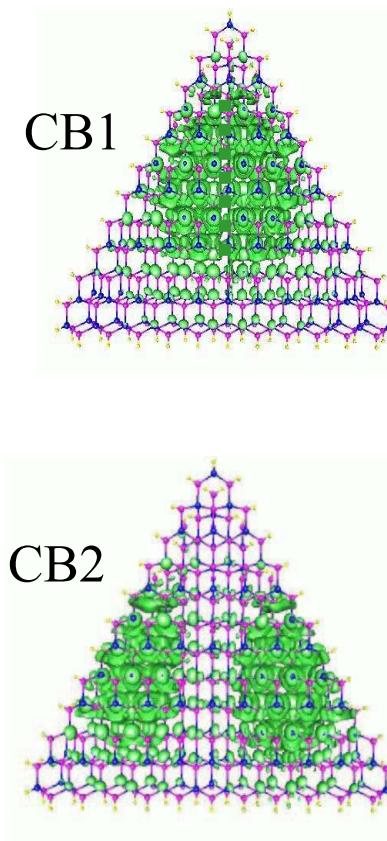
GaN (111) and (112) quantum wires (WZ)



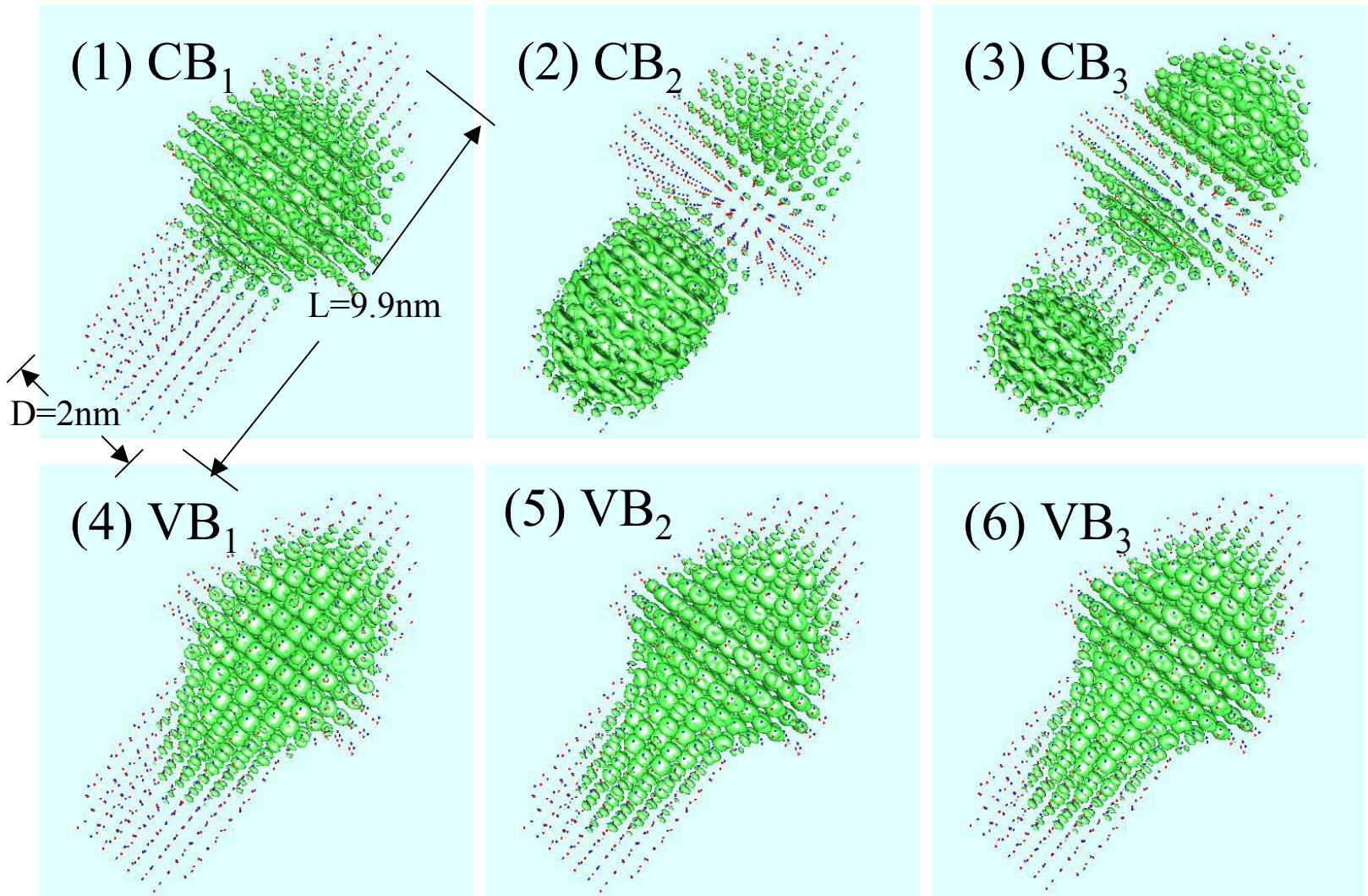
(111) GaN wire



(112) GaN wire

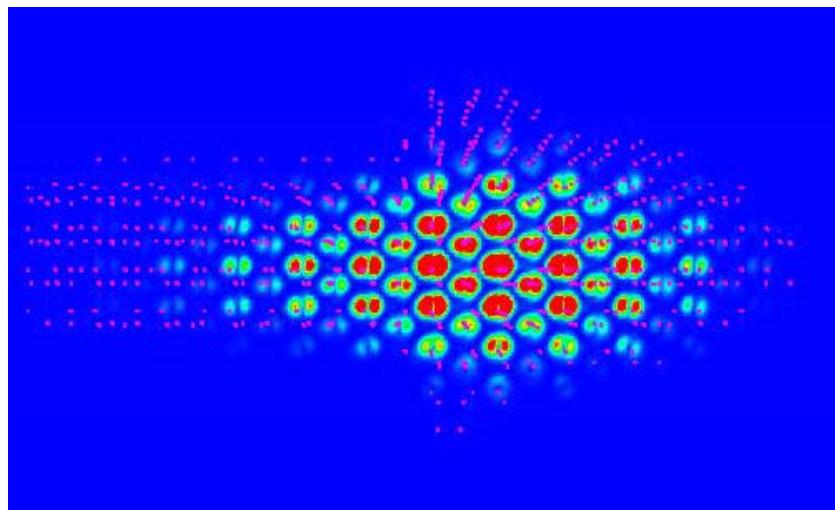


CdSe quantum dot: arrow shape

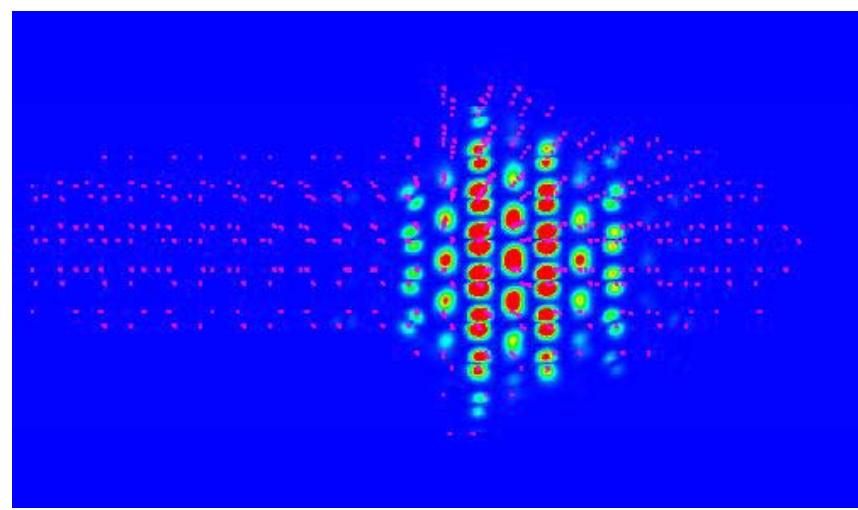


Different Bloch state characters for the VB states

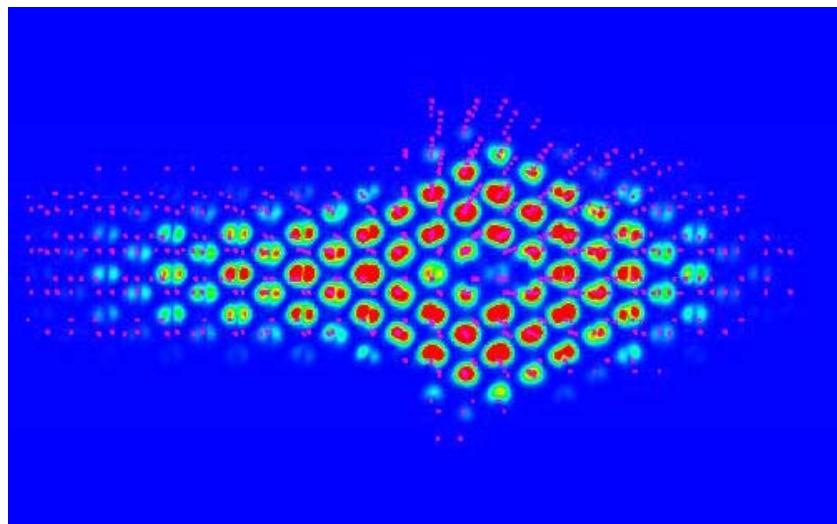
VB-1



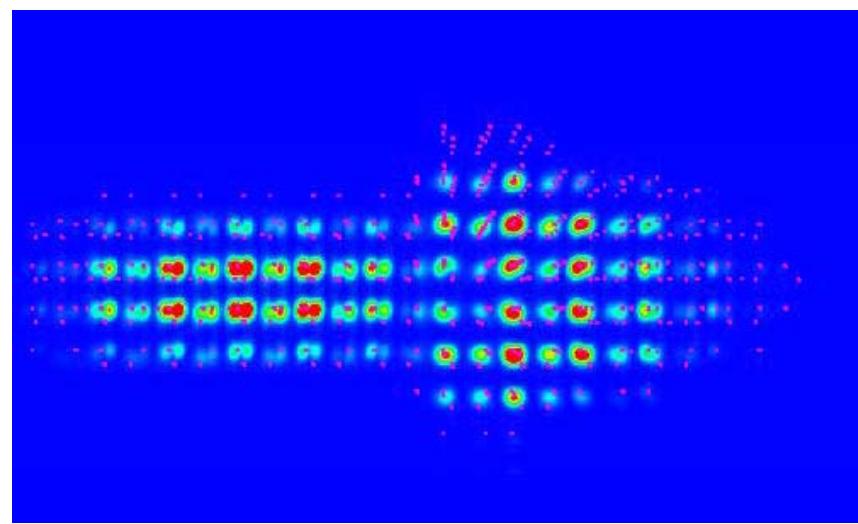
VB-2



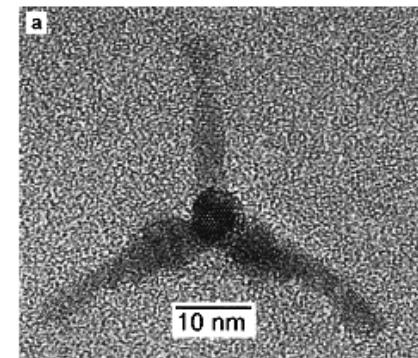
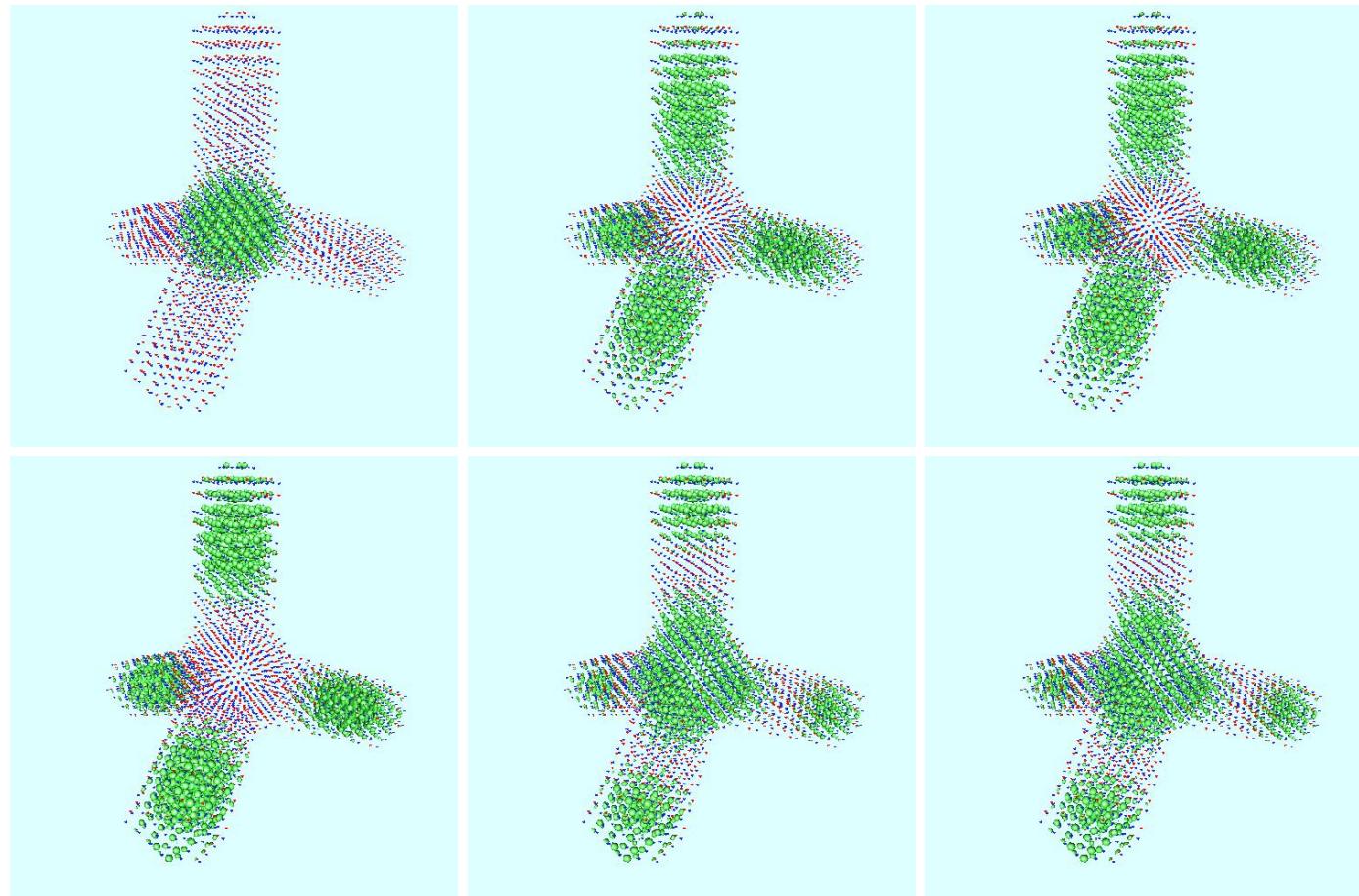
VB-3



VB-4

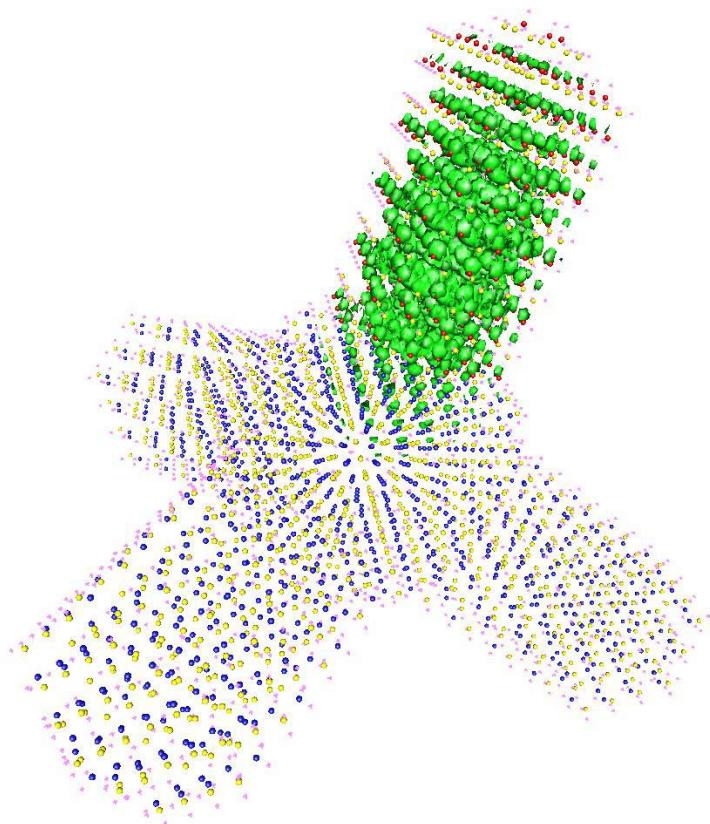


CdSe tetrapod electronic states

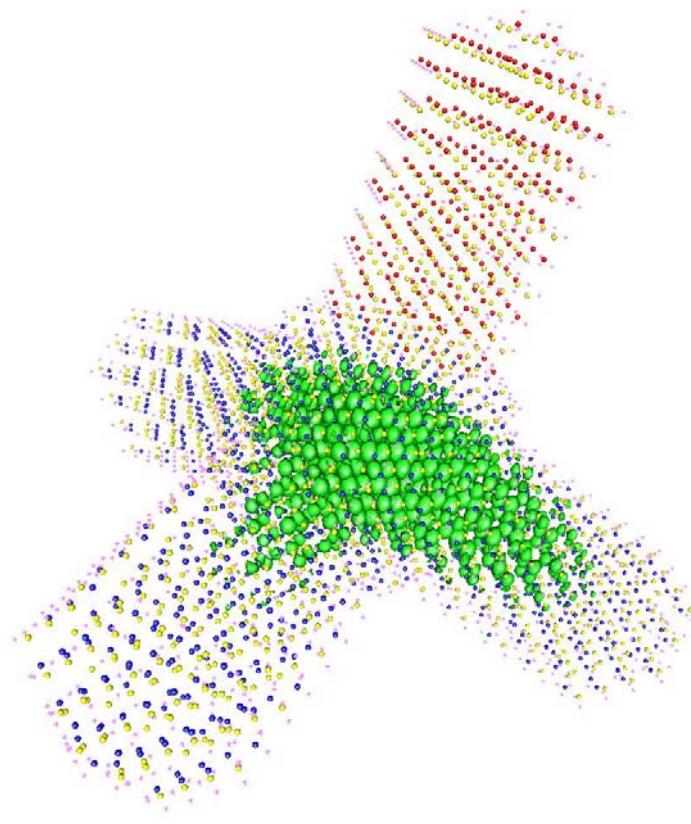


CdSe/CdTe tetrapod with one CdTe arm

Electron state

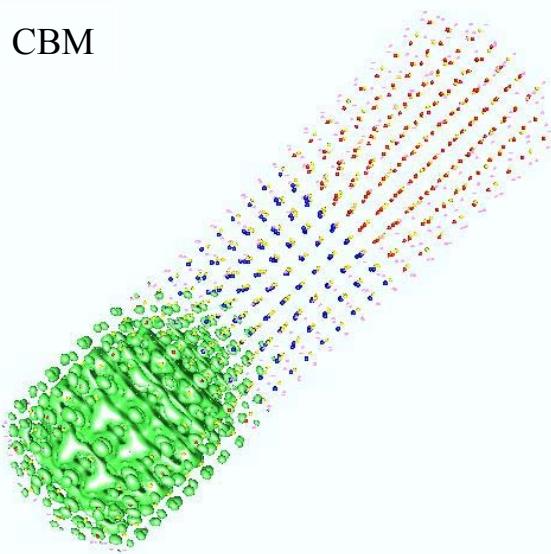


Hole state

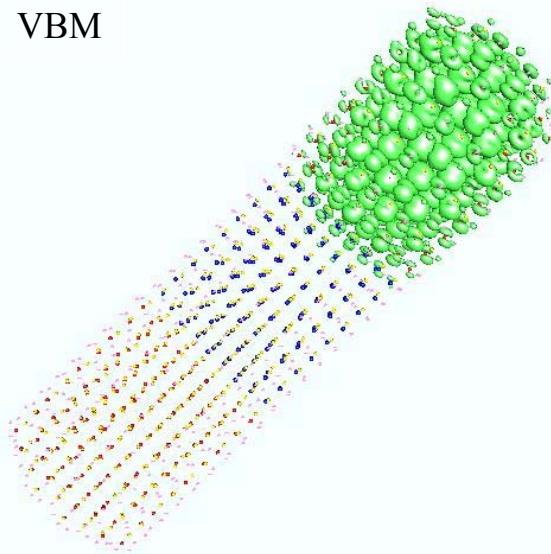


CdSe/CdS/CdSe quantum rod

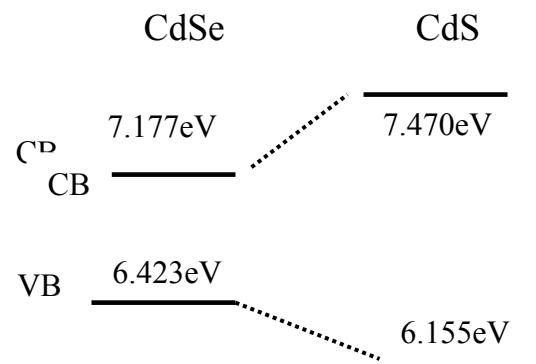
CBM



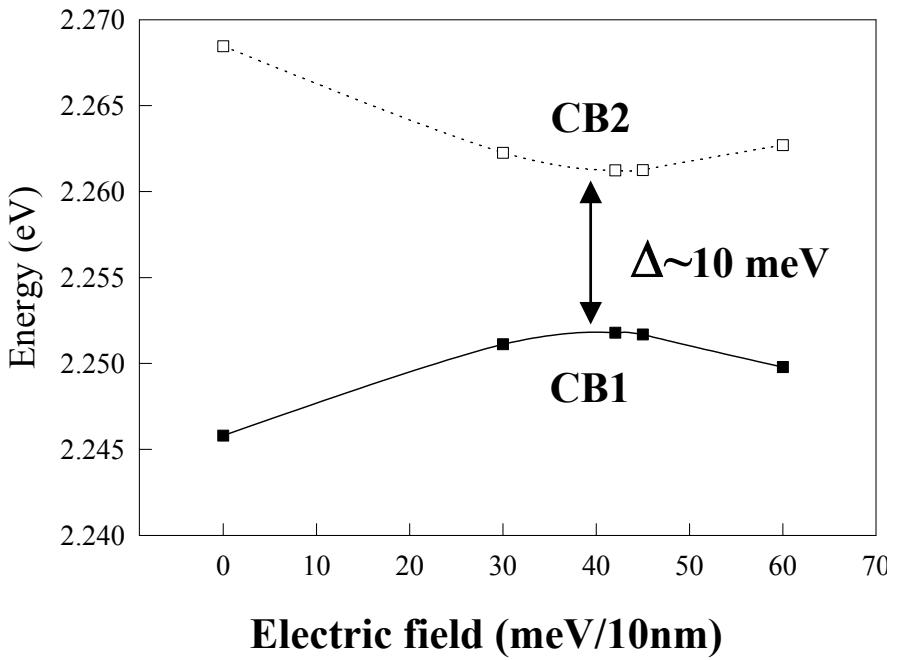
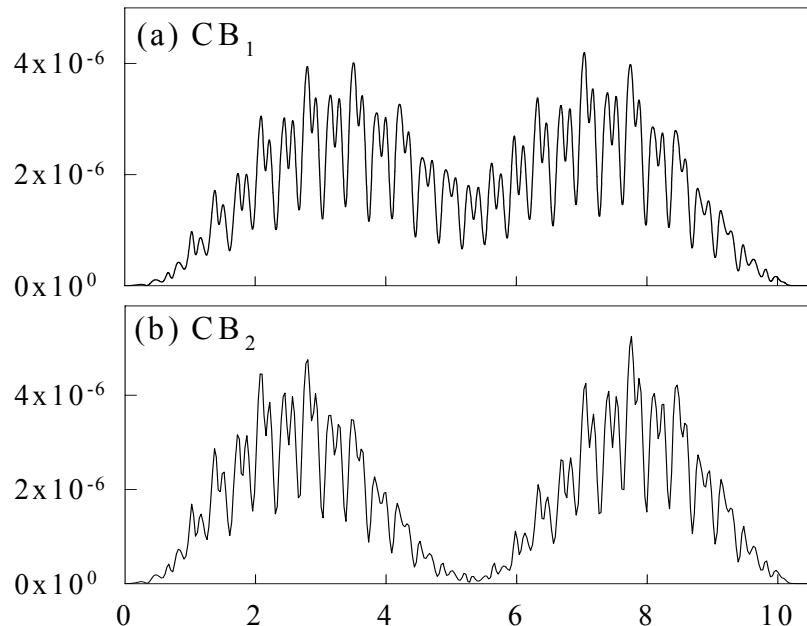
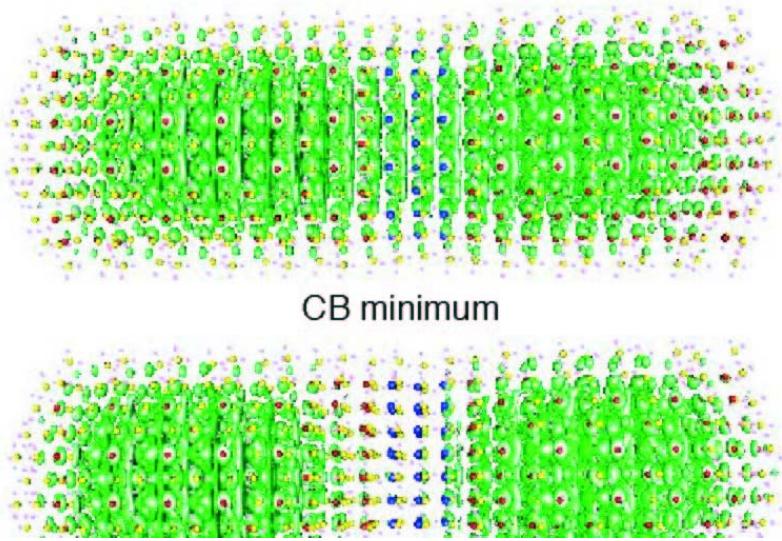
VBM



Band alignment of bulk CdSe/CdS



Anticrossing (coupling) states under electric field

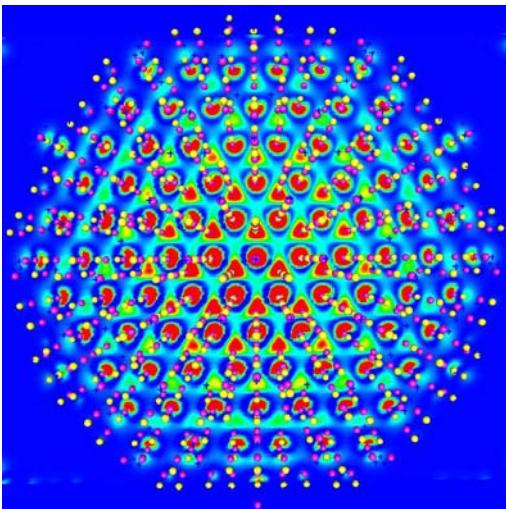


6 double layers of CdS: $\Delta=10$ meV
3 double layers of CdS: $\Delta=30$ meV

Core/shell quantum dots

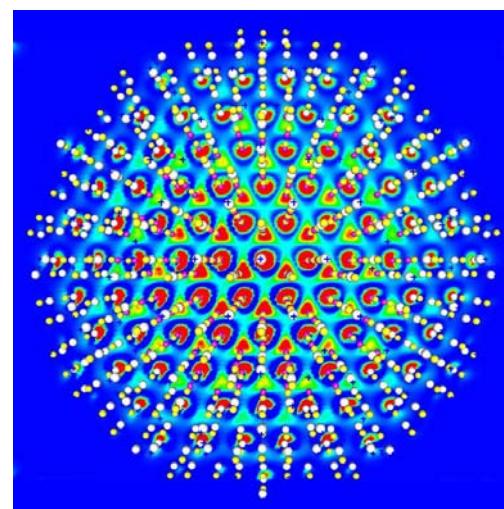
CdSe
CBM

CdSe

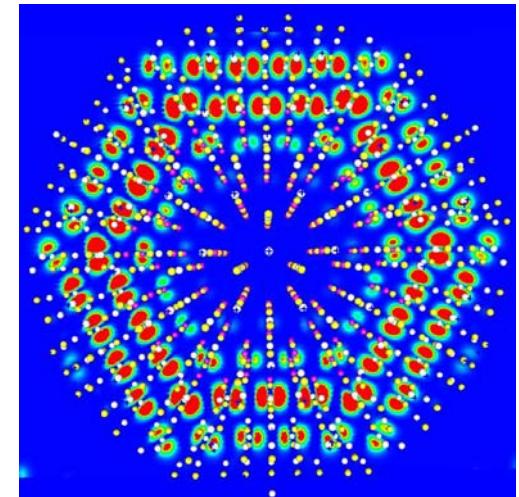
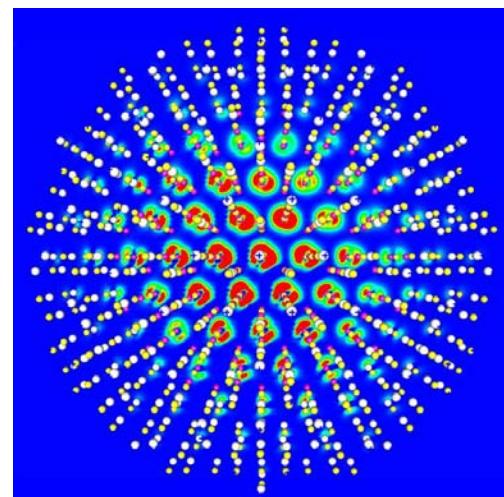
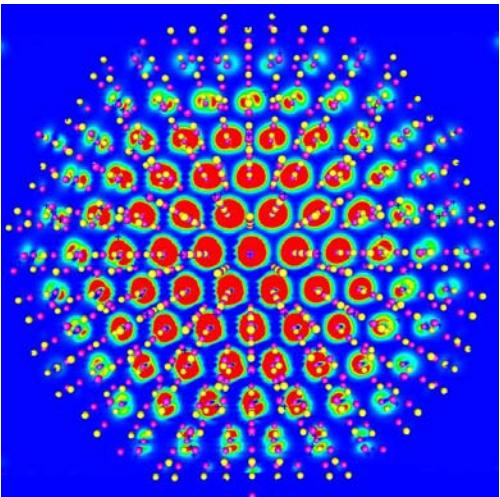
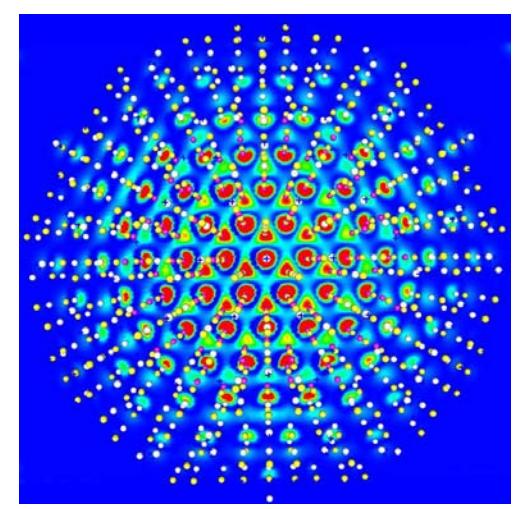


VBM

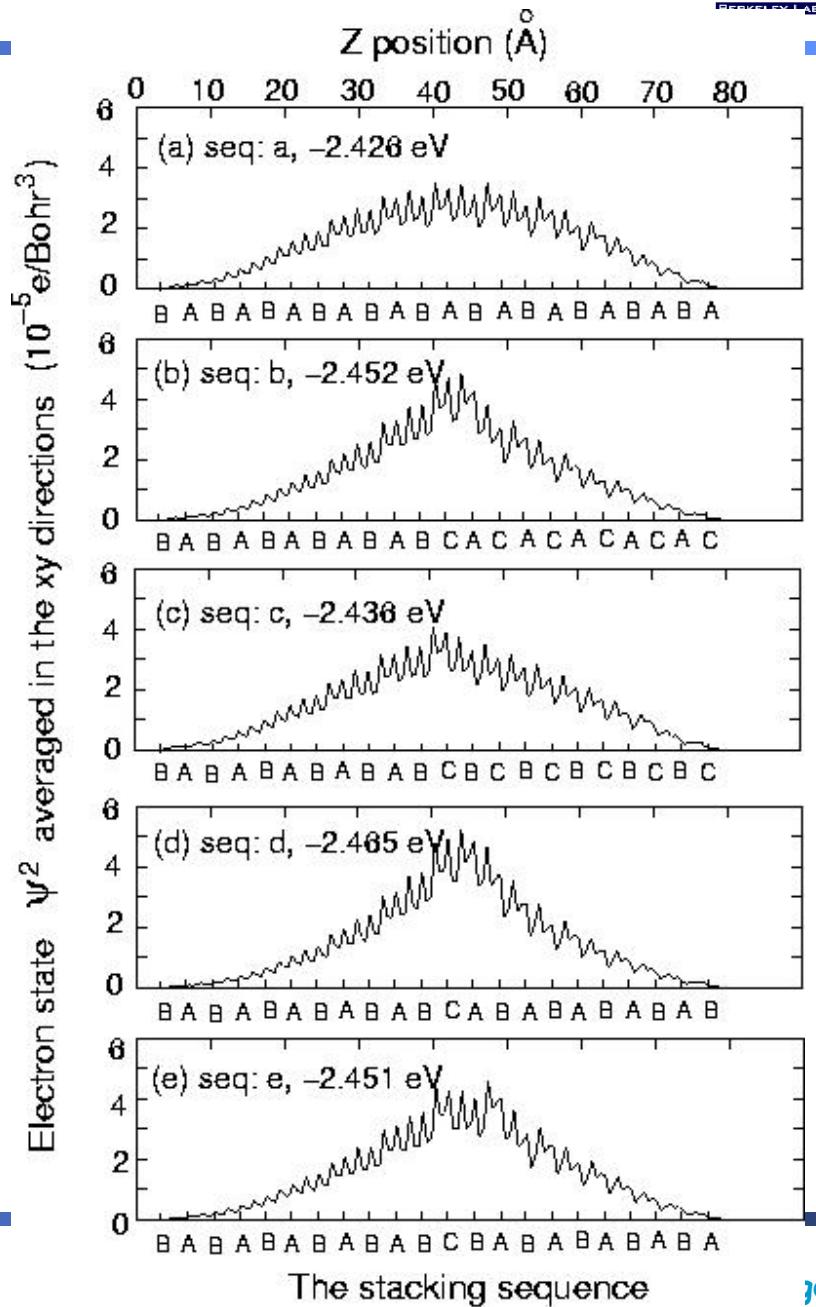
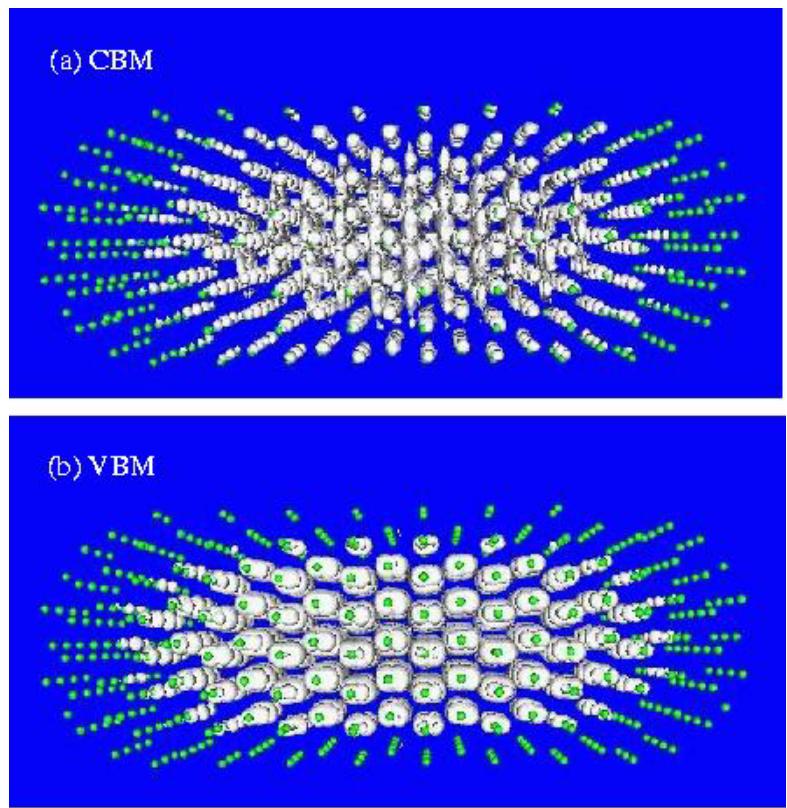
CdSe/CdS



CdSe/CdTe



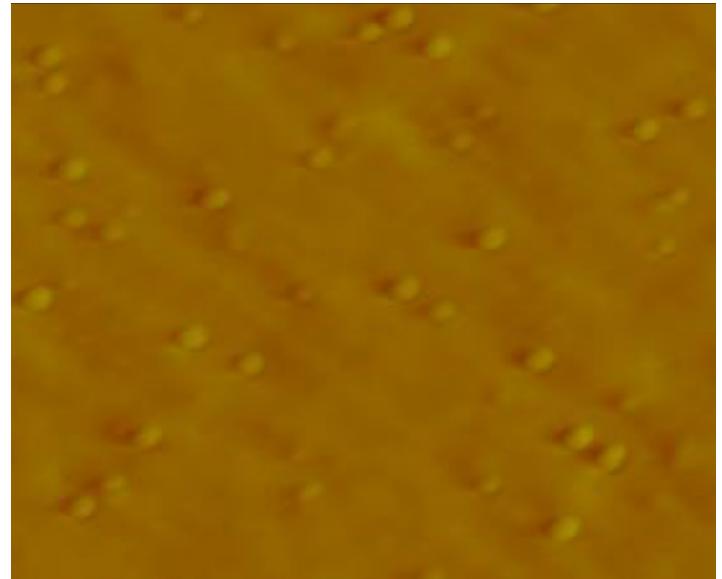
Effects of stacking faults



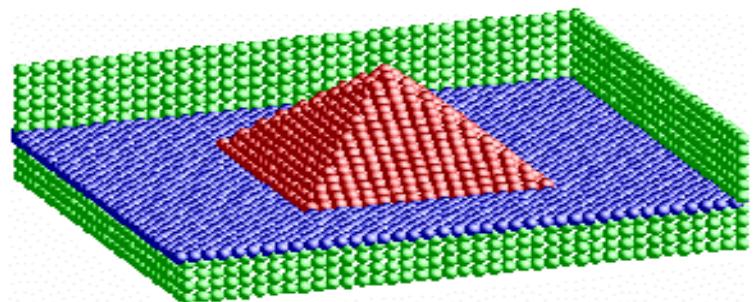
Self-assembled quantum dot

- Formed by themselves during MBE growth
- Strain between the dot and the substrate
- Size range ~ 100-500 Å,
~ a million atoms
- No dislocations, or surface defects
- Can be used for single electron device

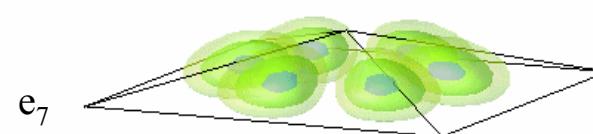
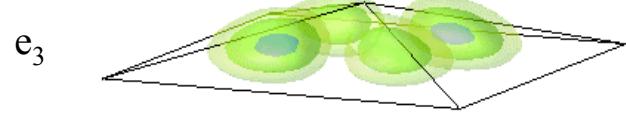
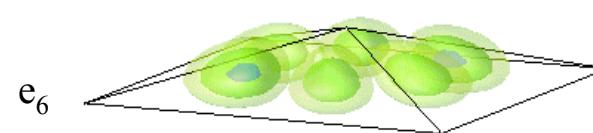
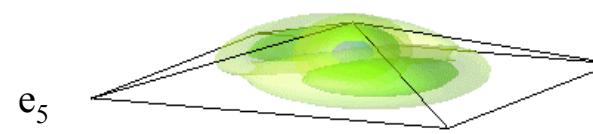
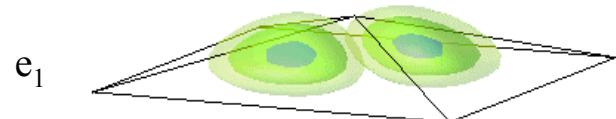
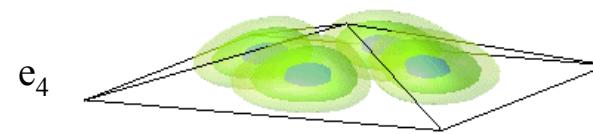
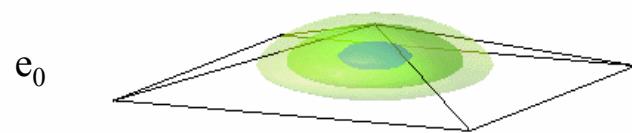
AFM image



InAs on GaAs substrate



Electronic states in embedded InAs quantum dot

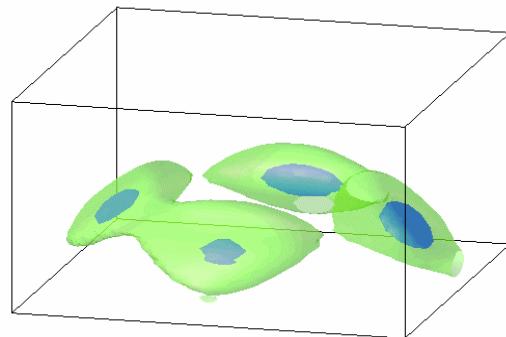


Hole states in embedded InAs quantum dots

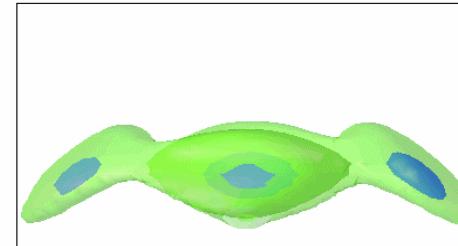
70h/200b InAs QD

$$\psi^2(h_0) + \psi^2(h_1) + \psi^2(h_2)$$

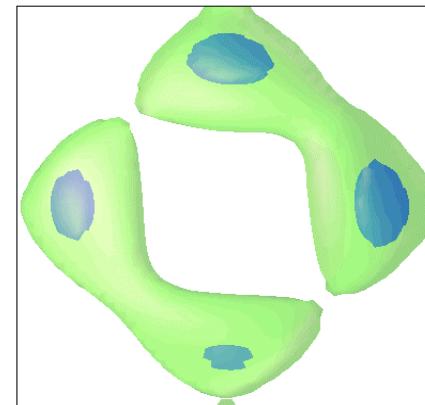
Angle



Side



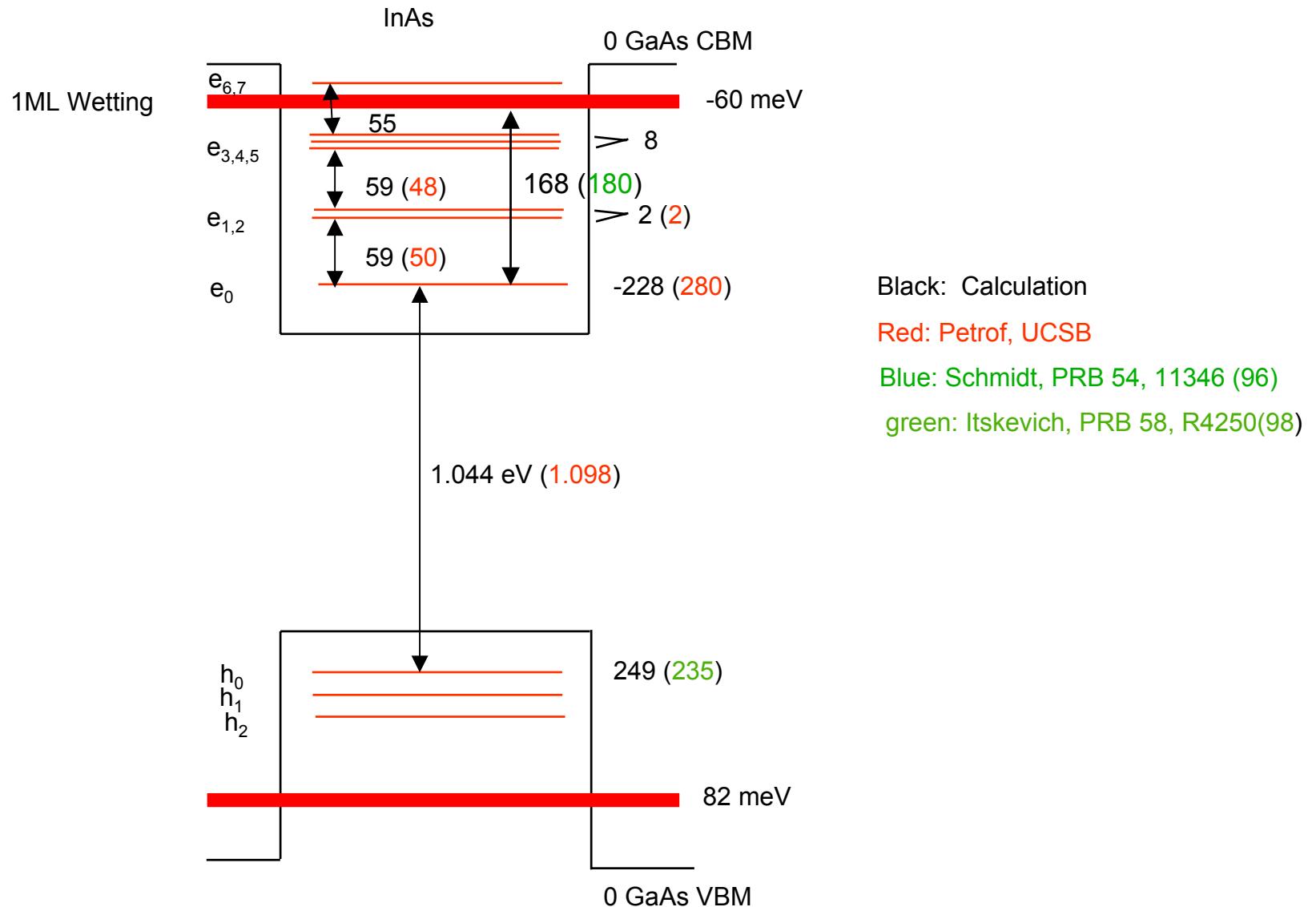
Top



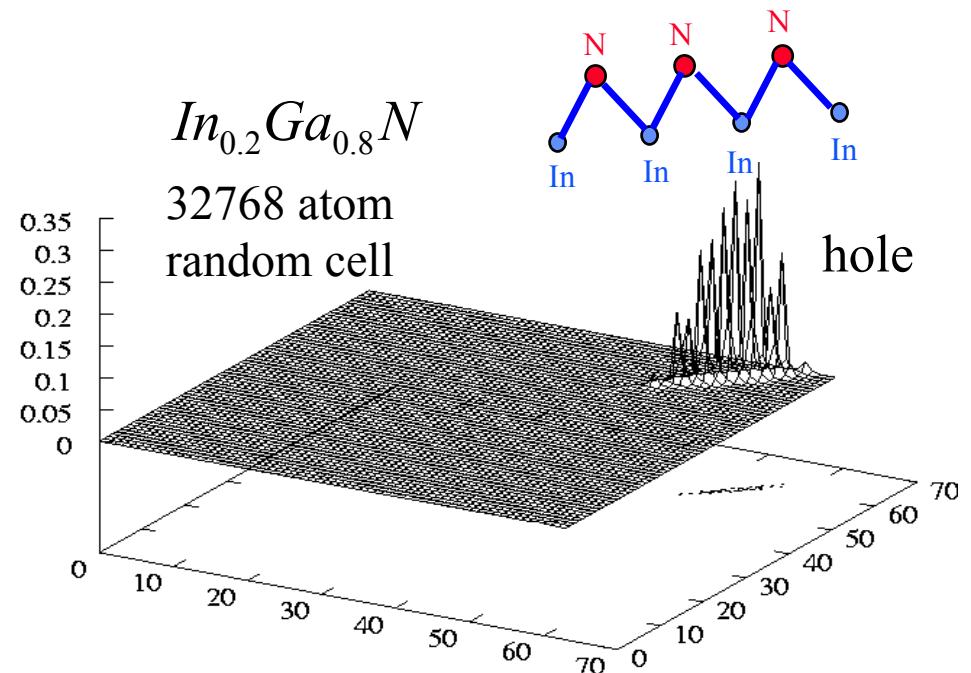
Green: 10%

Blue: 25%

Energy levels, comparison with experiment

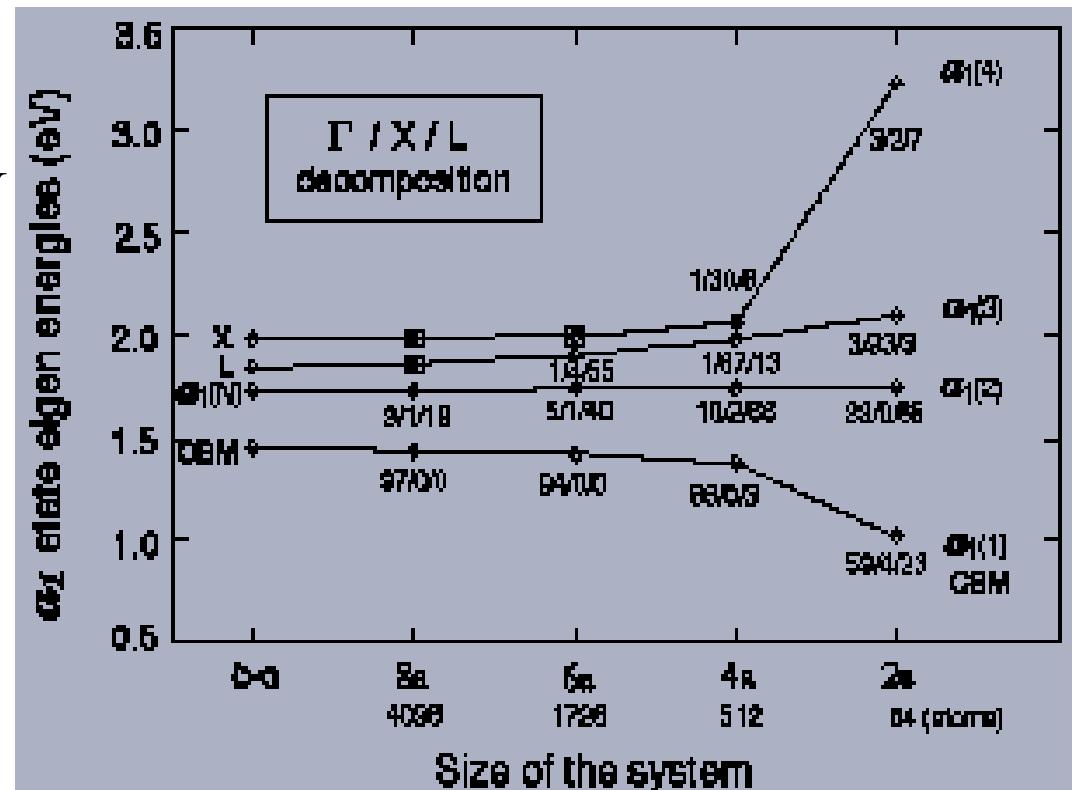
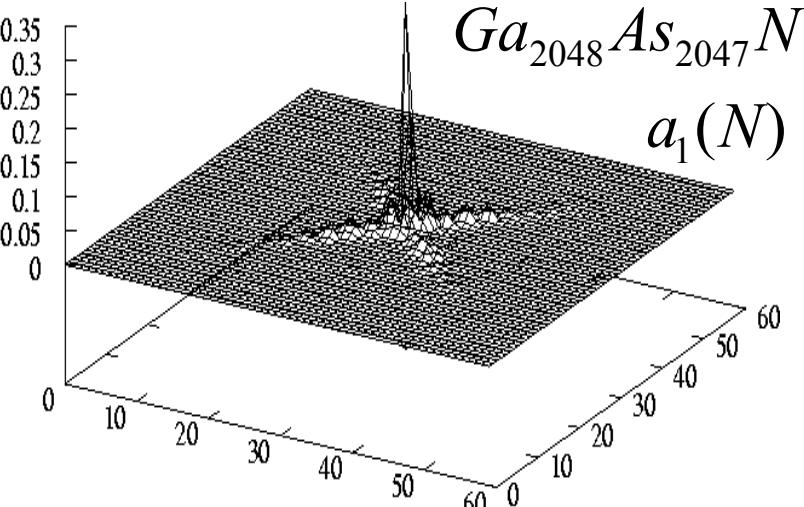


Hole localization in InGaN alloy

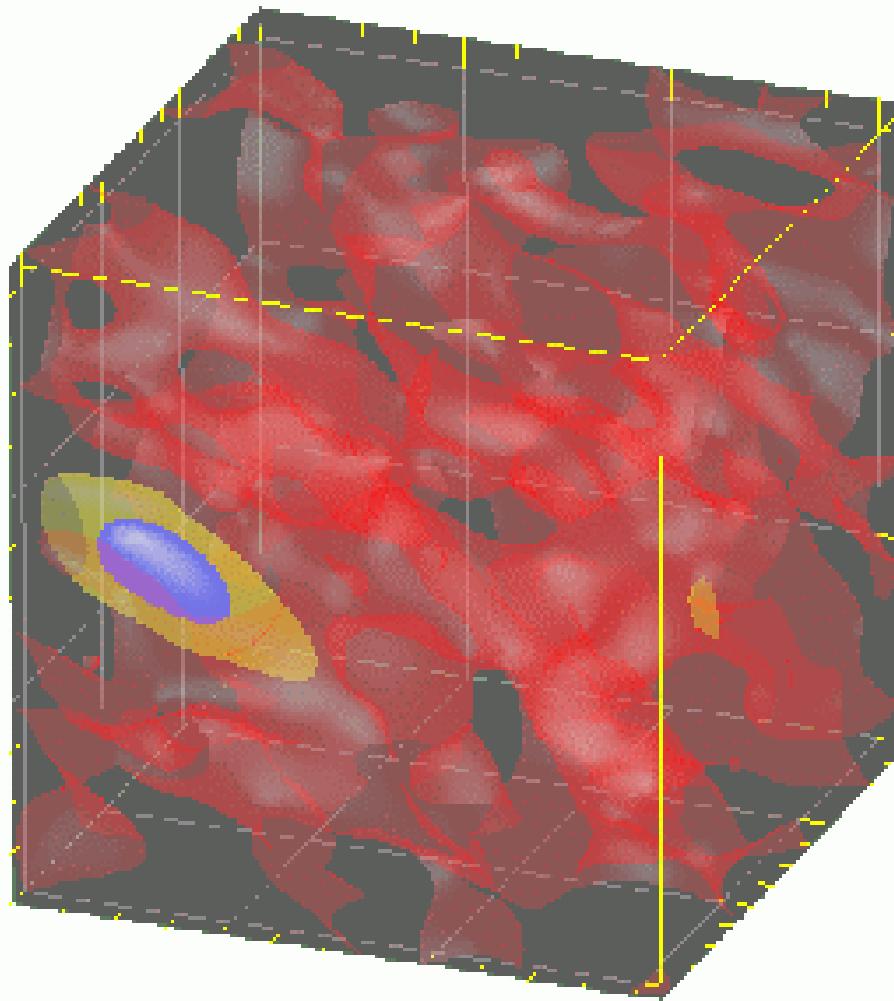


Blue Laser from InGaN

Impurity level calculation of GaAs:N



2 million atom GaAlAs alloy wavefunction



Conclusion

